Beyond local effective material properties for metamaterials

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(Received 1 June 2017; revised manuscript received 12 February 2018; published 26 February 2018)

To discuss the properties of metamaterials on physical grounds and to consider them in applications, effective material parameters are usually introduced and assigned to a given metamaterial. In most cases, only weak spatial dispersion is considered. It allows to assign local material properties, e.g., a permittivity and a permeability. However, this turned out to be insufficient. To solve this problem, we study here the effective properties of metamaterials with constitutive relations beyond a local response and take strong spatial dispersion into account. This research requires two contributions. First, bulk properties in terms of eigenmodes need to be studied. We particularly investigate the isofrequency surfaces of their dispersion relation are investigated and compared to those of an actual metamaterial. The significant improvement to effectively describe it provides evidence for the necessity to use nonlocal material laws in the effective description of metamaterials. Second, to be able to capitalize on such constitutive relations, also interface conditions need to be known. They are derived in this contribution for our form of the nonlocality using a generalized (weak) formulation of Maxwell's equations. Based on such interface conditions, Fresnel expressions are obtained that predict the amplitude of the reflected and transmitted plane wave upon illuminating a slab of such a nonlocal metamaterial. This all together offers the necessary means for the in-depth analysis of metamaterials characterized by strong spatial dispersion. The general formulation we choose here renders our approach applicable to a wide class of metamaterials.

DOI: 10.1103/PhysRevB.97.075439

I. INTRODUCTION

Assigning effective material properties is a main problem in the macroscopic description of optical metamaterials [1,2]. The desire of replacing such materials by hypothetical, homogeneous ones rises from the simplification when discussing them within a physical framework. Once a metamaterial is homogenized, it might be described and discussed on the same level as a natural material by its effective material properties. It can also be considered then in a plethora of applications. Exemplarily, we can mention perfect lenses [3], cloaks [4], broadband antireflection coatings [5,6], or directional antennas [7]; but there exist many more examples. Moreover, addressing numerically the wave propagation inside a homogeneous material is more efficient, i.e., less computational effort is required than in performing a rigorous computation of the full structure. However, it is of utmost importance that both descriptions for the same metamaterial, i.e., the actual and the homogenized metamaterial, provide the same response, up to a certain accuracy, to the electromagnetic field. Otherwise, the homogenization procedure is meaningless.

Various techniques have been established to assign effective material properties to optical metamaterials [8–18]. However, most previous techniques assume local constitutive relations, i.e., in a homogeneous material, the functional dependency of the auxiliary fields, **D** and **H**, is a linear combination of the macroscopic fields **E** and **B**, in which the coefficients are spatially independent [19,20],

$$\mathbf{D}(\mathbf{r},k_0) = \epsilon(k_0)\mathbf{E}(\mathbf{r},k_0) + \xi(k_0)\mathbf{B}(\mathbf{r},k_0)\,,$$
$$\mathbf{H}(\mathbf{r},k_0) = \mu^{-1}(k_0)\mathbf{B}(\mathbf{r},k_0) + \zeta(k_0)\mathbf{E}(\mathbf{r},k_0)\,, \qquad (1)$$

where $k_0 = \frac{\omega}{c_0}$. These equations are usually called local bianisotropic material laws, with effective material parameters ϵ, μ, ξ, and ζ and they are tensorial quantities, in general. A comprehensive review might be found in Ref. [21]. Although such constitutive relations are usually assumed, they exhibit several limitations [22–24]. In particular, the effective properties are only tensors that do not depend on the considered spatial frequency. However, they turn out to be inadequate when considering light propagation inside the metamaterial in an arbitrary direction and not just in the direction that was considered in the retrieval procedure [22,25]. Moreover, whereas it can be safely expected that such constitutive relations are valid when considering metamaterials for which the operational wavelength is much longer than the size of the unit cell, they fail to be predictive for most metamaterials that are operated in a regime where the wavelength is not much smaller than size of the unit cell but only smaller. Such operational regime, unfortunately, is necessary to observe many relevant dispersive effects.

To overcome these limitations, we propose in this work two formulations for advanced constitutive relations in order to

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model metamaterials beyond a local response and to take strong spatial dispersion into account. The applicability of these models is investigated. The justification for such models is derived by studying here the isofrequency surfaces of the dispersion relation obtained by our models that are compared to those of an actual metamaterial when considering the fundamental mode, i.e., the mode with the smallest imaginary part in the propagation constant. We find clear evidence that it is necessary to consider strong spatial dispersion, i.e., nonlocal constitutive relations in the effective description of the metamaterials.

To fully capitalize on such constitutive relations, we need to equip them with suitable interface conditions to describe the optical response from basic functional elements made from such metamaterials. Potentially, the simplest example for such functional element is a slab and we wish to know how light is reflected and transmitted from such a slab. Therefore, in this contribution, we also derive the associated interface conditions from first principles by relying on a generalized (weak) formulation of Maxwell's equations. These interface conditions allow to find the Fresnel equations for the reflection and transmission of a plane wave and for both polarizations from a slab. These Fresnel equations are also derived in this contribution.

In the context of nonlocal constitutive relations, comparable approaches were already employed to effectively homogenize metamaterials beyond their local regime [26–33]. In addition, considerable analytical work was employed in the study of nonlocal effects in plasmonic wire media based on cylindrical surface plasmon modes in Ref. [34] and on Mie scattering theory in Ref. [29]. However, very often they required a specific geometry for the metamaterial that motivated the formulation of specific constitutive relations. In contrast, in this work we propose a scheme based on a phenomenological *Ansatz* that can be applied to any periodic metamaterial made of subwavelength, resonant unit cells to describe homogenized metamaterials.

This paper is structured as follows. In Sec. II, we introduce our models from a very general *Ansatz*. Taking a specific fishnet metamaterial as an example, we apply in Sec. III our models to describe its dispersion relation and to show the improvement, compared to a dispersion relation derived with a local materials law. In Sec. IV, we make a basic statement on the formulation of the interface problem, the considered geometry, and add some remarks on the mathematical notation we have to put in place for discussing it. In Sec. V, we discuss the generalized solutions of Maxwell's equations for the respective constitutive relations. In Sec. VI, we derive the associated interface conditions and in Sec. VII, we outline the Fresnel equations to compute the reflection and transmission from a slab of such a homogenous metamaterial characterised by nonlocal constitutive relations. Discussion and conclusion of this work are contained in Sec. VIII.

II. CONSTITUTIVE RELATIONS

Let us consider a periodic metamaterial in which the inclusions are intrinsically nonmagnetic and reciprocal. We consider time-harmonic fields and only a linear response. It is therefore legitimate to assume that the electromagnetic response may be completely described by a nonlocal electric response, which can be written as the following integral form [35]:

$$\mathbf{D}(\mathbf{r},k_0) = \int_{\mathbb{R}^3} \hat{R}(\mathbf{r},\mathbf{r}',k_0)\mathbf{E}(\mathbf{r}',k_0)d\mathbf{r}' \tag{2}$$

and

$$\mathbf{H}(\mathbf{r},k_0) = \mathbf{B}(\mathbf{r},k_0)\,, \tag{3}$$

where the kernel $\hat{R}$ represents the electric response tensor, that spatially links in a nonlocal fashion the electric field $\mathbf{E}$ to the displacement field $\mathbf{D}$. In a homogeneous medium, the response kernel does not depend on the exact spatial position but only on the difference between two considered points in space. This suggests that the kernel $\hat{R}$ in Eq. (2) reduces to a difference kernel, i.e.,

$$\mathbf{D}(\mathbf{r},k_0) = \int_{\mathbb{R}^3} \hat{R}(\mathbf{r}-\mathbf{r}',k_0)\mathbf{E}(\mathbf{r}',k_0)d\mathbf{r}'\,, \tag{4}$$

which is a convolution integral. It is more convenient to formulate this constitutive relation in spatial Fourier space, such that the convolution becomes a product:

$$\tilde{\mathbf{D}}(\mathbf{k},k_0) = \tilde{\hat{R}}(\mathbf{k},k_0)\tilde{\mathbf{E}}(\mathbf{k},k_0)\,. \tag{5}$$

This expression is too general for practical purposes. In order to reach useful constitutive relations, we assume that the unit cells are subwavelength and the kernel can be expanded into a Taylor expansion with respect to the spatial frequency $\mathbf{k}$. Up to the fourth-order expansion, the kernel reads as the following:

$$\begin{aligned}\tilde{D}_i(\mathbf{k},k_0) = {}& (\delta_{ij}+a_{ij})\tilde{E}_j + b_{ijk}k_k\tilde{E}_j + c_{ijkl}k_kk_l\tilde{E}_j \\ & + d_{ijklm}k_kk_lk_m\tilde{E}_j + e_{ijklmn}k_kk_lk_mk_n\tilde{E}_j + \text{H.O.T.}\,,\end{aligned} \tag{6}$$

where Einstein's summation convention has been used. Note that all coefficients are in general complex functions of the frequency k_0. In the long-wavelength limit, i.e., $|\mathbf{k}| \to 0$, spatial dispersion disappears and the constitutive relation (6) reduces to the one known from an anisotropic medium with only an electric response in its local description, i.e.,

$$\tilde{D}_i(k_0) = (\delta_{ij}+a_{ij})\tilde{E}_j\,,$$

with the linear electric polarization $\tilde{P}_i = a_{ij}\tilde{E}_j$. Therefore the first line of Eq. (6) refers to the local permittivity of an anisotropic medium with

$$\epsilon_{ij}(k_0) = \delta_{ij} + a_{ij}(k_0)\,.$$

In order to determine the nature of the higher-order terms, we perform an inverse Fourier transform to real space. For plane waves, an inverse Fourier transform to the real space would lead to higher-order derivatives of the electric field with respect to spatial coordinates at the same position where the displacement field is to be evaluated, whereupon the constitutive relation becomes

$$\begin{aligned}D_i(\mathbf{r},k_0) = {}& (\delta_{ij}+a_{ij})E_j + b_{ijk}\partial_kE_j + c_{ijkl}\partial_{kl}E_j \\ & + d_{ijklm}\partial_{klm}E_j + e_{ijklmn}\partial_{klmn}E_j + \text{H.O.T.}\,,\end{aligned} \tag{7}$$

where all coefficients were multiplied by a prefactor $(-\mathrm{i})^n$, with n being the order of the spatial derivative. To be able to practically work with feasible constitutive relation, particular in the context of metamaterials, assumptions to these coefficients have to be made in order to reach well established effective medium theories that shall describe the actual metamaterial. For instance, a frequently made assumption is that

$$b_{ijk}(k_0)\partial_k \stackrel{!}{=} [\xi(k_0)\nabla\times]_{ij} \tag{8}$$

and/or

$$c_{ijkl}(k_0)\partial_{kl} \stackrel{!}{=} [\nabla\times\alpha(k_0)\nabla\times]_{ij}\,. \tag{9}$$

If these assumptions are met indeed and it is furthermore assumed that all higher-order terms vanish, local constitutive relations identical to those in Eq. (1) can be derived by exploiting degrees of freedom that allow to suitably gauge Maxwell's equations. The first assumption leads to a local optical activity (gyrotropy), whereas the second assumption leads to a local magnetic permeability [21], respectively. Consequently, the permeability becomes dispersive and reads

$$\mu(k_0) = \left[\mathbb{I} - k_0^2\alpha(k_0)\right]^{-1}, \tag{10}$$

where $\mathbb{I}$ is the identity matrix in three dimensions. We will refer to these assumptions as the weak spatial dispersion (WSD) approximation. "Spatial dispersion" because it results form nonlocal material laws and "weak" because there is a transformation that gives local constitutive relations, with a local magnetoelectric coupling and a local permeability, hence a local bi-anisotropic medium. However, it has been already shown that these assumptions are not enough to adequately describe the dispersion relation of an actual metamaterial beyond the paraxial regime [22]. In a principal coordinates system, dispersion relations obtained from WSD are either of the hyperbolic kind, if the principal components of the permittivity or the permeability have opposite signs, or of the elliptic kind otherwise. However, most metamaterials with a nonlocal response usually show dispersion relations that widely differ from hyperbolas or ellipsoids. Exemplarily, we study in the following one example for a metamaterial where the dispersion relation of the fundamental mode indeed differs from that obtained within WSD. This suggests that the WSD approximation is not enough and we, therefore, need to go beyond it and have to retain higher-order terms in the expansion. Here, we retain up to the fourth spatial-derivative of the electric field in Eq. (7).

In order to proceed, we assume WSD as a starting point and extend it into two directions. In our first model, we extend WSD by a, quite similar, but fourth-order law in which the fourth-order expansion coefficients take the following form:

$$e_{ijklmn}(k_0)\partial_{klmn} \stackrel{!}{=} [\nabla\times\nabla\times\gamma(k_0)\nabla\times\nabla\times]_{ij}\,, \tag{11}$$

where $\gamma = \mathrm{diag}(\gamma_x,\gamma_y,\gamma_z)$ is a diagonal matrix and represents a higher-order material parameter. This special form is chosen due to mathematical reasons. The coefficients that obey Eq. (11) do not change the variational class of Maxwell's equations and hence, interface conditions can be found by means of variational methods as described further below. Even higher-order terms again are assumed to vanish.

Our second model remains to be a second-order law but it strictly orients on symmetry considerations of a unit cell of a specific metamaterial. Specifically, it takes more coefficients into account than those that lead to local constitutive relations, i.e., assumptions (8) and (9). The model takes every coefficient into account as required by the symmetry of the considered metamaterial. The tensor elements of the kernel in Eq. (5) reflect the spatial symmetry of the actual structure. The degrees of freedom that the kernel might have can be reduced considerably depending on the symmetry of metamaterials. In this approach, we do not include fourth-order terms in expansion (6), as it already gives significant improvements compared to WSD. The treatment of both approaches simultaneously, i.e., retaining both unit cell's symmetry conditions and a fourth-order response, in principle, can be applied but the analysis becomes much more involved and is beyond the scope of this paper. In both models and for the sake of simplicity, we only consider metamaterials whose unit cells have a center of symmetry. By the presence of this (spatial inversion-)symmetry, all odd terms in expansion (7) vanish, hence no gyrotropic medium or optical activity is considered. However, it has to be stressed that this is not a general limitation. It is done here to concentrate only on one specific aspect.

A. Analysis by retaining fourth-order response

Let us now investigate the dispersion relation and the consequences that arise from the following material law:

$$\tilde{\mathbf{D}}(\mathbf{k},k_0) = \epsilon\tilde{\mathbf{E}} - \mathbf{k}\times(\alpha\mathbf{k}\times\tilde{\mathbf{E}}) + \mathbf{k}\times\mathbf{k}(\times\gamma\mathbf{k}\times\mathbf{k}\tilde{\mathbf{E}})\,. \tag{12}$$

For convenience, we assume that the coordinate system of the laboratory corresponds to the coordinate system of the principle axis of the metamaterial. The material parameters are therefore diagonal and read

$$\epsilon = \begin{pmatrix}\epsilon_x & 0 & 0\\ 0 & \epsilon_y & 0\\ 0 & 0 & \epsilon_z\end{pmatrix},\quad \alpha = \begin{pmatrix}\alpha_x & 0 & 0\\ 0 & \alpha_y & 0\\ 0 & 0 & \alpha_z\end{pmatrix},\quad \gamma = \begin{pmatrix}\gamma_x & 0 & 0\\ 0 & \gamma_y & 0\\ 0 & 0 & \gamma_z\end{pmatrix}. \tag{13}$$

This is a direct extension of WSD by the next higher-order response such that Maxwell's equations remain of the variational class. Hence interface conditions may be found by means of variational methods. It has to be noted that this model is by itself not unique. The additional fourth-order term, together with the standard second-order term, can be reformulated as a nonlocal magnetic response with respect to (w.r.t.) the reformulation in Ch. XII of Ref. [35] and the gauge transformation in Ch. 2 of Ref. [21]. In the anisotropic case, the expression for the permeability is more complicated than before the reformulation, which renders the latter pointless. However, nice expression can be obtained if the material parameters are scalars, i.e., when the model considers an isotropic material. The resulting magnetic response is then

$$\mu(k_0,\mathbf{k}) = \frac{1}{1 - k_0^2[\alpha(k_0) + \mathbf{k}^{\mathrm{T}}\cdot\mathbf{k}\ \gamma(k_0)]}\,.$$

In general, the dispersion relation can be derived by solving the wave-type equation

$$\mathbf{k} \times \mathbf{k} \times \tilde{\mathbf{E}} + k_0^2 \tilde{\mathbf{D}}[\tilde{\mathbf{E}}] = 0 \,. \tag{14}$$

This represents three coupled equations of second degree for the spatial frequency $\mathbf{k}$. Under the assumption that $k_x = 0$ and considering the z direction as the principle propagation direction, the dispersion relation for the transversal electric (TE) mode can be found. The dispersion relation expresses here the functional dependency of the frequency and the wave-vector components. In TE-polarization, the mode has a nonzero electric field component normal to the k_y-k_z plane. The dispersion relation reads as

$$k_y^2 \mu_y + k_z^2 \mu_z - \left(k_y^2 + k_z^2\right)^2 \gamma_x \mu_y \mu_z k_0^2 = \epsilon_x \mu_y \mu_z k_0^2 \,,$$

where according to Ref. [21] $\mu_i = \frac{1}{1 - k_0^2 \alpha_i}$. The solutions are

$$k_z^2(k_y, k_0) = -k_y^2 + p_0^{\mathrm{TE}} \pm \sqrt{\left(p_0^{\mathrm{TE}}\right)^2 - q_1^{\mathrm{TE}} + 2k_y^2\left(p_1^{\mathrm{TE}} - p_0^{\mathrm{TE}}\right)} \tag{15}$$

with the frequency-dependent coefficients

$$\begin{aligned} q_1^{\mathrm{TE}}(k_0) &= \frac{\epsilon_x(k_0)}{\gamma_x(k_0)} \,, \\ p_0^{\mathrm{TE}}(k_0) &= \left[2k_0^2 \gamma_x(k_0) \mu_y(k_0)\right]^{-1} , \\ p_1^{\mathrm{TE}}(k_0) &= \left[2k_0^2 \gamma_x(k_0) \mu_z(k_0)\right]^{-1} , \end{aligned} \tag{16}$$

where q_1^{TE} has the dimension of m^{-4} and both p_0^{TE} and p_1^{TE} the dimension of m^{-2}. For the transversal magnetic (TM) mode, i.e., the mode that has an electric field in the k_y-k_z plane, we obtain

$$\begin{aligned} & k_y^2 \epsilon_y + k_z^2 \epsilon_z - k_y^2 k_z^2 (\epsilon_y \gamma_y + \epsilon_z \gamma_z) \mu_x k_0^2 \\ & \quad - \left(k_y^4 \epsilon_y \gamma_y + k_z^4 \epsilon_z \gamma_z\right) \mu_x k_0^2 = \epsilon_y \epsilon_z \mu_x k_0^2 \,, \end{aligned}$$

with the solutions

$$k_z^2(k_y, k_0) = -\frac{1}{2}\left(q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}\right) k_y^2 + p_0^{\mathrm{TM}} \pm \sqrt{\left[p_0^{\mathrm{TM}} + \frac{q_0^{\mathrm{TM}} - q_1^{\mathrm{TM}}}{2} k_y^2\right]^2 - p_1^{\mathrm{TM}}} \,, \tag{17}$$

where

$$\begin{aligned} q_0^{\mathrm{TM}}(k_0) &= \frac{\epsilon_y(k_0)}{\epsilon_z(k_0)} \,, \\ q_1^{\mathrm{TM}}(k_0) &= \frac{\gamma_z(k_0)}{\gamma_y(k_0)} \,, \\ p_0^{\mathrm{TM}}(k_0) &= \left[2k_0^2 \mu_x(k_0) \gamma_y(k_0)\right]^{-1} , \\ p_1^{\mathrm{TM}}(k_0) &= \frac{\epsilon_y(k_0)}{\gamma_y(k_0)} \,. \end{aligned} \tag{18}$$

Here, both q_0^{TM} and q_1^{TM} are dimensionless, while p_0^{TM} and p_1^{TM} have the dimensions of m^{-2} and m^{-4}, respectively. We would also like to note that this model embraces previous models for strong spatial dispersion relations that were specifically derived for the plasmonic wire medium. For example, it can reproduce the results [Eq. (10)] obtained from Ref. [29], if we choose for the TM mode the coefficients $(q_0^{\mathrm{TM}}, q_1^{\mathrm{TM}}, p_0^{\mathrm{TM}}, p_1^{\mathrm{TM}})$ properly. This finding also means that our model is directly applicable to describe nonlocal effects in wire media but goes beyond that as at the level of the material parameters more degrees of freedom exist.

Due to the higher number of degrees of freedom, the functional dependency of $k_z(k_y, k_0)$ is more advanced, in comparison to the previously proposed relation for a material with local constitutive relations (WSD) that reads as [22]

$$k_z^{\mathrm{WSD}^2}(k_y, k_0) = \alpha_1(k_0) + \alpha_2(k_0) k_y^2 \,. \tag{19}$$

Here, the coefficients α_1 and α_2 depend on the polarization where for the TE mode they read

$$\begin{aligned} \alpha_1^{\mathrm{TE}}(k_0) &= k_0^2 \epsilon_x(k_0) \mu_y(k_0) \,, \\ \alpha_2^{\mathrm{TE}}(k_0) &= -\frac{\mu_y(k_0)}{\mu_z(k_0)} \,, \end{aligned} \tag{20}$$

and

$$\begin{aligned} \alpha_1^{\mathrm{TM}}(k_0) &= k_0^2 \epsilon_y(k_0) \mu_x(k_0) \,, \\ \alpha_2^{\mathrm{TM}}(k_0) &= -\frac{\epsilon_y(k_0)}{\epsilon_z(k_0)} \,, \end{aligned} \tag{21}$$

for the TM mode. In general, isofrequency contours of media described by Eq. (19) are limited to two cases: they are either of hyperbolic or elliptic kind. This limitation is lifted by introducing more complicated dispersion relations, e.g., our fourth-order response. To illustrate the possible features an isofrequency contour may have when considering such fourth-order constitutive relations, Fig. 1 shows some plots of $k_z(k_y)$ for a fixed frequency k_0 for some generically chosen parameters. The obtained isofrequency contours give rise to more advanced curves. They allow for a homogeneous description of a metamaterial with dispersive features inaccessible by a local material. It is also of imperative importance to mention that multiple solutions to the wave equation also means that multiple plane waves exist as solutions for a given pair or frequency and transverse wave-vector components. This is clearly different to the case of weak spatial dispersion, where only a single plane wave exists as solution. Due to the linearity of the wave equation [Eq. (14)], the eigenmode is a superposition of four plane waves:

$$\begin{aligned} \mathbf{E}(\mathbf{k}, \omega) = {} & \mathbf{E}_0^{++} e^{\mathrm{i}(\mathbf{k}^{++}\mathbf{r} - \omega t)} + \mathbf{E}_0^{+-} e^{\mathrm{i}(\mathbf{k}^{+-}\mathbf{r} - \omega t)} \\ & + \mathbf{E}_0^{-+} e^{\mathrm{i}(\mathbf{k}^{-+}\mathbf{r} - \omega t)} + \mathbf{E}_0^{--} e^{\mathrm{i}(\mathbf{k}^{--}\mathbf{r} - \omega t)} \,. \end{aligned} \tag{22}$$

This effect is always associated with nonlocal material laws. Furthermore, it can be seen from Fig. 1 (and later from Fig. 2) that for every set of parameters (q_0, q_1, p_0, p_1), i.e., every subfigure, the eigenmodes are usually attenuated differently. In the limiting case where the nonlocal parameter $\gamma \to 0$, one of the eigenmodes has an eigenvalue k_z whose imaginary part goes to infinity while the other mode tends to the solution known from WSD. In this contribution, we focus on the fundamental mode, i.e., the mode with the smallest $\Im(k_z)$, and show that this is already leading to significant improvements to the current situation.

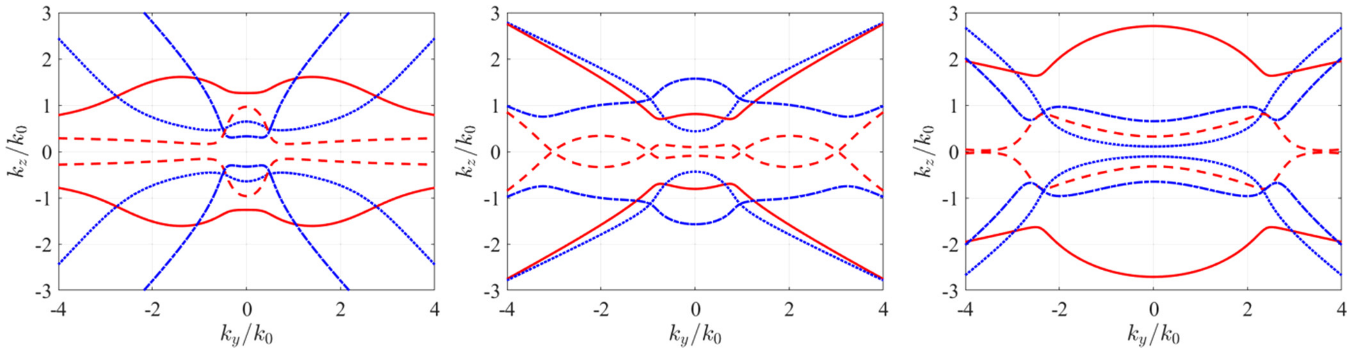

FIG. 1. Examples of isofrequency contours decomposed into real (red) and imaginary (blue) parts for the inner + solutions (solid red and dotted blue lines) as well as for the inner − solutions (dashed red and dash-dot blue lines) of Eqs. (15) and (17). The left figure shows the isofrequency contours of the TE mode for $p_0^{\mathrm{TE}} = (1 + 0.5\mathrm{i})\ \mu\mathrm{m}^{-2}$, $p_1^{\mathrm{TE}} = (-1.6 - 1.5\mathrm{i})\ \mu\mathrm{m}^{-2}$, and $q_1^{\mathrm{TE}} = (2 + 0.6\mathrm{i})\ \mu\mathrm{m}^{-4}$. The center and the right figures show the isofrequency contours of the TM modes for $p_0^{\mathrm{TM}} = (-1 - 0.5\mathrm{i})\ \mu\mathrm{m}^{-2}$, $p_1^{\mathrm{TM}} = (-3.1 + 0.1\mathrm{i})\ \mu\mathrm{m}^{-4}$, $q_0^{\mathrm{TM}} = -3.1 + 0.5\mathrm{i}$ and $q_1^{\mathrm{TM}} = 3 + 0.5\mathrm{i}$, and $p_0^{\mathrm{TM}} = (3.5 - 0.5\mathrm{i})\ \mu\mathrm{m}^{-2}$, $p_1^{\mathrm{TM}} = (-11.1 + 0.1\mathrm{i})\ \mu\mathrm{m}^{-4}$, $q_0^{\mathrm{TM}} = -2.1 + 0.1\mathrm{i}$ and $q_1^{\mathrm{TM}} = 3 + 0.5\mathrm{i}$, respectively.

B. Analysis with respect to structure's symmetry

In this model, instead of taking higher orders in the expansion of the kernel into account, we take a deeper look in the geometry of an actual (real) structure. As an example we consider the fishnet metamaterial (see Fig. 3) as a subject of homogenization. The unit cell of the fishnet metamaterial is symmetric under transformation with respect to three orthogonal mirror planes, i.e., the permittivity distribution obeys

$$\epsilon(x,y,z) = \epsilon(\pm x, \pm y, \pm z)\,.$$

This symmetry class is also known as orthorhombic symmetry [36], noted as $\mathrm{D}_{2\mathrm{h}}$. According to this consideration, we can write the expansion of the displacement field in the form

$$\tilde{\mathbf{D}}(\mathbf{k},k_0) = \epsilon\tilde{\mathbf{E}} - \mathbf{k}\times(\alpha\mathbf{k}\times\tilde{\mathbf{E}}) - \sum_{j\in\{x,y,z\}} k_j(\beta^j k_j\tilde{\mathbf{E}}). \quad (23)$$

Moreover, additionally, we assume that the coordinate system of the laboratory corresponds to the coordinate system of the principle axis of the metamaterial. Therefore the material tensors are considered to be diagonal. In this coordinate system, the nonlocal material properties read as

$$\beta^x = \begin{pmatrix}\beta_x & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & 0\end{pmatrix},\quad \beta^y = \begin{pmatrix}0 & 0 & 0\\ 0 & \beta_y & 0\\ 0 & 0 & 0\end{pmatrix},$$
$$\beta^z = \begin{pmatrix}0 & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & \beta_z\end{pmatrix}. \quad (24)$$

Please note the indices of β. If the index j is used as a superscript to β, it refers to a dyadic in the proper direction, i.e., a matrix. If the index j is used as a subscript to β, it refers to a scalar. The dyadic, for example, reads as $\beta^j = \beta_j\hat{\mathbf{e}}_j\hat{\mathbf{e}}_j$,

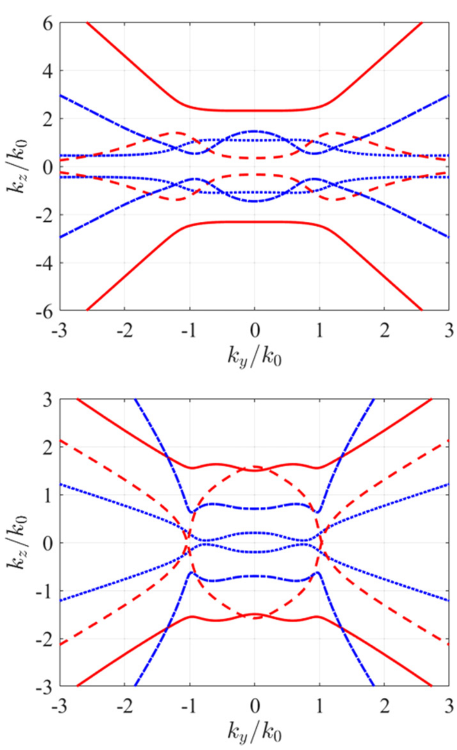

FIG. 2. Examples of isofrequency contours decomposed into real (red) and imaginary (blue) parts for the inner + solutions (solid red and dotted blue lines) as well as for the inner − solutions (dashed red and dash-dot blue lines) of Eq. (30). The upper figure shows the isofrequency contours for $p_0^{\mathrm{TM}} = (-1 + 0.5\mathrm{i})\ \mu\mathrm{m}^{-2}$, $p_1^{\mathrm{TM}} = 2.1 + 0.1\mathrm{i}$, $q_0^{\mathrm{TM}} = (2.1 + 2.5\mathrm{i})\ \mu\mathrm{m}^{-2}$, and $q_1^{\mathrm{TM}} = (-2 + 0.5\mathrm{i})\ \mu\mathrm{m}^{-2}$, while the bottom one for $p_0^{\mathrm{TM}} = (1 + 1.11\mathrm{i})\ \mu\mathrm{m}^{-2}$, $p_1^{\mathrm{TM}} = -1.1 + \mathrm{i}$, $q_0^{\mathrm{TM}} = (1.1 - 0.3\mathrm{i})\ \mu\mathrm{m}^{-2}$, and $q_1^{\mathrm{TM}} = (-0.2 + 2.5\mathrm{i})\ \mu\mathrm{m}^{-2}$.

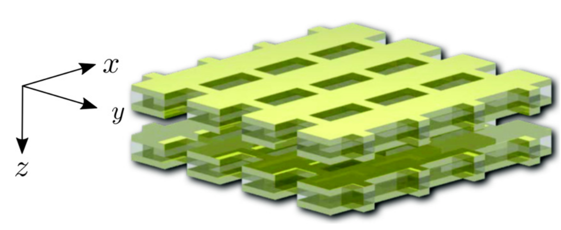

FIG. 3. Fishnet metamaterial consisting of a biperiodic structure with periods $\Lambda_x = \Lambda_y = 600$ nm and $\Lambda_z = 200$ nm with rectangular holes with the width $w_y = 100$ nm and $w_x = 316$ nm. It compromises a stack of layers made of two 45-nm Ag layers separated by a thin dielectric spacer, 30 nm of MgF_2. The remaining space is filled with air.

where $\hat{\mathbf{e}}_j$ is a unit vector in j direction. More importantly, some of the second-order terms may be written in a similar way as in the WSD. The only difference to the WSD is the higher-order susceptibility contribution β_i that couples k_i^2 with E_i. This term, however, cannot be set to zero *a priori* for a metamaterial with the considered symmetry of the fishnet. It has to be taken into account and appears on the same footing as the other second-order terms. These other terms in k^2 are assumed to obey the condition that yields a local magnetic response, i.e., Eq. (9). With these assumptions, the model will always have a nonlocal electric response, in spite of the reformulation proposed in Ref. [35] or the gauge transformation in Ref. [21], as $\beta_i k_i^2$ couples longitudinally to the electric field. This results to the same permeability in Eq. (10). The dispersion relation follows from solving the wave equation as described in Eq. (14). We assume equally that $k_x = 0$ and we study a plane wave with a wave vector in the k_y-k_z plane. The dispersion relation for the TE mode is then

$$k_z^2\mu_z + k_y^2\mu_z = \mu_z\mu_z\epsilon_x k_0^2 , \tag{25}$$

with the solutions

$$k_z^2(k_y,k_0) = p_0^{\mathrm{TE}} + q_0^{\mathrm{TE}} k_y^2, \tag{26}$$

where

$$q_0^{\mathrm{TE}}(k_0) = -\frac{\mu_y}{\mu_z} , \tag{27}$$

$$p_0^{\mathrm{TE}}(k_0) = \epsilon_x\mu_y k_0^2 . \tag{28}$$

The fact that the TE mode does not experience any strong spatial dispersion relies on the nature of the expansion in Eq. (23) with the nonlocal response that couples only in the direction of the electric field, hence no cross coupling between the displacement field and the electric field as can be seen in Eq. (23). In contrast, a more complicated dispersion relation is found for the TM modes with

$$\begin{aligned} &k_z^2(\epsilon_z + \beta_z\epsilon_y\mu_x k_0^2) + k_y^2(\epsilon_y + \beta_y\epsilon_z\mu_x k_0^2) \\ &- k_z^2k_y^2\beta_z\beta_y\mu_x k_0^2 - k_z^4\beta_z - k_y^4\beta_y = \epsilon_y\epsilon_z\mu_x k_0^2 , \end{aligned} \tag{29}$$

where according to Ref. [21] $\mu_i = \frac{1}{1-k_0^2\alpha_i}$. This equation is biquadratic in k_y and k_z and describes more complicated isofrequencies than quadratic equations, e.g., given by WSD. Of course, the limit $(\beta_y,\beta_z) \to (0,0)$ restores the dispersion relation given by WSD. The solutions of Eq. (29) are

$$\begin{aligned} k_z^2(k_y,k_0) &= p_1^{\mathrm{TM}} k_y^2 + q_0^{\mathrm{TM}} + p_0^{\mathrm{TM}} \\ &\pm \sqrt{\left(p_1^{\mathrm{TM}}k_y^2 + p_0^{\mathrm{TM}} - q_0^{\mathrm{TM}}\right)^2 + 2q_1^{\mathrm{TM}}\left(\frac{p_1^{\mathrm{TM}}}{p_0^{\mathrm{TM}}}k_y^4 + k_y^2\right)} \end{aligned} \tag{30}$$

with the related coefficients that read

$$q_0^{\mathrm{TM}}(k_0) = \frac{\epsilon_z(k_0)}{2\beta_z(k_0)} ,$$

$$p_0^{\mathrm{TM}}(k_0) = \frac{k_0^2}{2}\epsilon_y(k_0)\mu_x(k_0) ,$$

$$q_1^{\mathrm{TM}}(k_0) = \frac{\epsilon_y(k_0)}{2\beta_z(k_0)} ,$$

$$p_1^{\mathrm{TM}}(k_0) = -\frac{k_0^2}{2}\beta_y(k_0)\mu_x(k_0) . \tag{31}$$

It has to be noted that all coefficients have the dimension of m^{-2}, except p_1 being dimensionless. Here, we have four independent coefficients, which increase the number of degrees of freedom to four. Previously, e.g., in Eq. (19), the dispersion relation contained only two independent coefficients, α_1 and α_2, hence only two degrees of freedom. Moreover, this holds for both strong spatial dispersion (SSD) models, each of the Eqs. (30), (15), and (17) yield four possible solutions for $k_z(k_y,k_0)$ from which two with positive and two with negative imaginary parts. We consider here only solutions with a positive imaginary part as they describe exponentially damped solutions in our principle propagation direction. In contrast, the two solutions with a negative imaginary part would correspond to exponentially growing solutions. They are unphysical for a passive medium. However, this still suggests that in the actual homogeneous medium characterized by material laws beyond WSD, more than a single mode is excited at the interface where the continuity of the tangential wave-vector components dictates which modes are excited. To simplify the analysis, we concentrate on investigating the fundamental mode, i.e., the mode with the smallest positive imaginary part. In general, the imaginary parts of the two solutions with $\Im(k_z) > 0$ may cross and a mode transition has to be taken into account. Here as well, Fig. 2 shows some isofrequency contours from selected parameter sets. The complexity of their shapes suggests the ability to capture the effects of SSD. In the next section, we show the importance of retaining these nonlocal effects in the effective description of the metamaterial by directly showing the improvements that follow from taking SSD into account, as introduced in Eqs. (12) and (23).

III. APPLICATION TO A FISHNET METAMATERIAL

In this section, a numerical experiment is done to get access to the dispersion relation of an actual structure as a reference. We consider the fishnet metamaterial shown in Fig. 3 as an example, which is known to exhibit a negative refractive index at optical frequencies. The geometrical parameters are taken from literature [37]. The unit cell's dimensions are $\Lambda_x = \Lambda_y = 600$ nm and $\Lambda_z = 200$ nm. The rectangular holes are made of perpendicularly aligned nanowires with thicknesses of $w_y = 100$ nm and $w_x = 316$ nm. It is made of two 45-nm silver (Ag) layers where their permittivity obey the Drude model for metals, with the plasma frequency being $\omega_p = 13700$ THz and the relaxation rate $\Gamma = 85$ THz. These layers are separated by a 30-nm magnesium fluoride (MgF_2) spacer whose permittivity is assumed to be nondispersive ($\epsilon_{\mathrm{MgF}_2} = 1.9044$) in the frequency range of interest. Furthermore, the unit cell is symmetric with respect to spatial inversion, i.e., $\epsilon(\mathbf{r}) = \epsilon(-\mathbf{r})$.

In order to calculate the Bloch mode dispersion relation, i.e., $k_z = k_z(k_x,k_y,k_0)$ where k_0 represents the wave number in free space, a plane-wave expansion *Ansatz* is numerically performed. In general, k_z can be complex. Its real part, $\Re(k_z)$, refers to the oscillatory part while its imaginary part, $\Im(k_z)$,

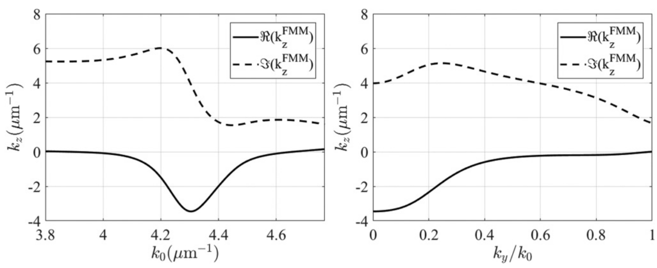

FIG. 4. (Left) Dispersion relation of the fundamental mode for $k_x = k_y = 0$ calculated by a plane-wave expansion *Ansatz* using the Fourier modal method (FMM) algorithm. It shows a resonance around $k_0 = 4.3\ \mu\text{m}^{-1}$ in which $\Re(k_z) < 0$. (Right) Isofrequency contour in the xz plane at the resonance wave number.

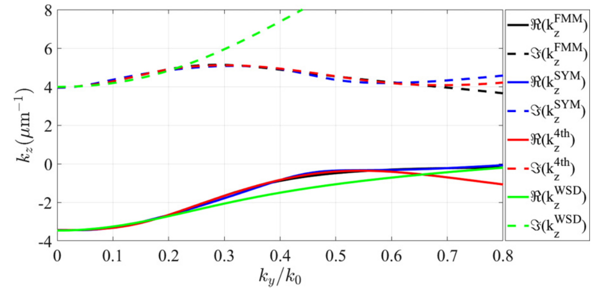

FIG. 5. Isofrequency contours $k_z = k_z(k_y)$ at the resonance frequency of the fishnet corresponding to a wave number of $k_0 = 4.3\ \mu\text{m}^{-1}$. Solid (dashed) curve represents the real (imaginary) part. The blue (crosses) curves are obtained from fitting Eq. (30) to the reference curve (black). It shows a good agreement up to $k_y = 0.3k_0$. The red (bullets) curves are obtained from fitting Eq. (17) to the reference curve and show a good agreement up to $k_y = 0.4k_0$. Meanwhile, the green (diamonds) curves, which are obtained from WSD, are showing only an agreement within the paraxial regime, i.e., for $k_y < 0.1k_0$.

denotes the energy loss in the principal propagation direction. We restrict our considerations to the solutions with $\Im(k_z) > 0$. The dominating Bloch mode, i.e., the fundamental Bloch mode that prevails after a finite propagation length is the one with the smallest positive $\Im(k_z)$ as all higher modes experience much stronger damping. Figure 4 shows both dispersion relation and isofrequency contours of the fundamental Bloch mode for different transverse vectors $k_y \geqslant 0$ at a fixed frequency and for different frequencies at a fixed transverse wave vector, respectively. Here we restrict ourselves to the k_y-k_z plane, i.e., $k_x = 0$. The mode is TM polarized. For wave numbers around $4.3\ \mu\text{m}^{-1}$, we observe negative $\Re(k_z)$, which implies a negative index, i.e., momentum and energy flux propagate in opposite directions. We are basically interested in homogenizing the material around this resonance wave number. Figure 4 (right) shows the isofrequency contour for $k_0 = 4.3\ \mu\text{m}^{-1}$ of the real structure. This numerically obtained isofrequency contour of the actual metamaterial has to be reproduced by the dispersion relation of the effective medium from WSD as well as from both SSD with some fixed set of parameters. The comparison is based on a least absolute deviations fit by optimizing the parameters (q_0, p_0, q_1, p_1) of fundamental TM modes from the SSD models and (α_1, α_2) from the WSD. As a quantity of measure, we define the merit function as

$$\delta(k_0) = \frac{\sum_{k_y} \left| 1 - \frac{k_z^{\mathrm{i}\,2}(k_y,k_0)}{k_z^{\mathrm{FMM}\,2}(k_y,k_0)} \right| w(k_y)}{\sum_{k_y} w(k_y)}, \tag{32}$$

with a suitably defined weight function $w(k_y)$. Here, $\mathrm{i} \in \{\text{WSD, 4th, SYM}\}$ denotes the model taken into account. Fourth and SYM refer to the fourth-order and to the symmetry model, respectively. As all the expressions were derived from a Taylor expansion for small $\mathbf{k}$ [see Eq. (6)], it is legitimate to introduce a weight $w(k_y)$ such that the fitting procedure is more focused for small k_y. Here we chose an exponentially decreasing dependency, i.e.,

$$w(k_y) = \mathrm{e}^{-\alpha k_y}, \tag{33}$$

where α was chosen to be $\alpha = 2.5\Lambda_y$, with $\Lambda_y = 0.6\ \mu$m being the lateral period of the fishnet structure. The results for a selected frequency—here we chose the worst case scenario, i.e., at the resonance—when considering the optimized parameters are depicted in Fig. 5. It shows the isofrequency contour in both the real and imaginary part of the dispersion relation numerically calculated for the actual fishnet metamaterial and the dispersion relation obtained for the best fit of parameters at the resonance frequency for the three different models considered. The parameter set of the SSD models for the best fit and the right signs are summarized in Table I. For the parameters of WSD, i.e., relation (19), we obtain $\alpha_1^{\mathrm{TM}} = (-5.85 - 28.12\mathrm{i})\ \mu\text{m}^{-2}$ and $\alpha_2^{\mathrm{TM}} = -2.49 + 3.79\mathrm{i}$. Clearly, WSD is only in a good agreement with the isofrequency contour of the real structure in the paraxial regime, i.e., $k_y \ll k_0$. Beyond the paraxial regime, we recognize from the shape of the black curves of $k_z(k_y)$, that WSD, which by nature is either an ellipse or a hyperbola, is not enough to describe the dispersion relation of such complicated structure. This limitation can be lifted by considering nonlocal constitutive relations as proposed above. The actual dispersion relation can be much better described when homogenizing the metamaterial with constitutive relations beyond the local model. To quantify the actual improvement, we study the merit function as a function of the frequency for the three different constitutive relations obtained in here. Each value for the merit function has been obtained from an individual fit at a specific frequency.

Figure 6 shows the improvements in effectively describing the metamaterial with nonlocal constitutive relations for all

TABLE I. Parameter set of the SSD models for the best fit as shown in Fig. 5. Depending on the model there is always one sign [the ± sign in Eqs. (17) and (30)] that fits better to the fundamental Bloch mode. Bear in mind that parameters from different models have different expressions and dimensions.

Model and sign:	4th(−)	SYM(+)
p_0^{TM}	$(-1.01 - 56.62\mathrm{i})\ \mu\text{m}^{-2}$	$(-1.97 - 13.85\mathrm{i})\ \mu\text{m}^{-2}$
p_1^{TM}	$(-2.35 + 0.28\mathrm{i})\times 10^3\ \mu\text{m}^{-2}$	$-7.21 - 1.12\mathrm{i}$
q_0^{TM}	$0.804 + 0.807\mathrm{i}$	$(-18.2 - 10.61\mathrm{i})\ \mu\text{m}^{-2}$
q_1^{TM}	$-26.43 - 21.80\mathrm{i}$	$(-27.7 + 33.95\mathrm{i})\ \mu\text{m}^{-2}$

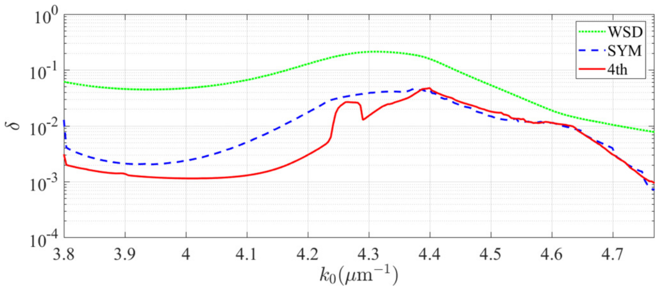

FIG. 6. Deviations from reference (FMM) in logarithmic scale. Over all frequencies, modeling metamaterials with nonlocal material laws makes more sense for a realistic homogenization.

of the simulated frequencies. Both SSD models are more accurate than WSD. The integrated error, which expresses how good a specific constitutive relation in a homogenized medium can explain the actual dispersion relation of the given metamaterial, is in average two orders of magnitude better for the nonlocal material laws. In resonance, the deviation is strongest irrespective of the considered model. However, this is somehow expected that the effective description tends to be inaccurate in the resonance regime. Nevertheless, the findings immediately imply that retaining nonlocal constitutive relations is required for a more realistic homogenization of metamaterials.

IV. MAXWELL'S EQUATIONS AND THE CONSIDERED CONSTITUTIVE RELATIONS

Now we have been settling the important finding that considering these nonlocal constitutive relation improves the effective description of metamaterials. This was exemplified at the fishnet geometry. We have been discussing in particular the dispersion relation of the eigenmodes and showed that we can capture their dispersive nature much better. These eigenmodes are a fundamentally important concept in eigenmodes, derived from the fact that an arbitrary field can always be expanded into a superposition of eigenmodes; of course just in a linear system. Having analytical access to these dispersion relations allows to evolve the fields in the bulk. However, this ability to describe the propagation of light in the bulk is not yet the entire story. Instead, we also need to know how an external field coming from an exterior medium can excite these eigenmodes. To answer this question requires to know the respective interface conditions. For a local material characterized by only a weak spatial dispersion, the derivation of the interface conditions is a straightforward task. It leads to the conclusion that the tangential electric and magnetic field **E** and **H** and the normal component of the electric displacement and the magnetic induction **D** and **B** are required to be continuous. These four interface conditions are just enough to fix all unknown quantities when solving, e.g., for the amplitudes of the reflected and transmitted plane waves at an interface in both polarizations, i.e., TE and TM. That is the case because for the WSD medium only a single plane wave is allowed by the dispersion relation, i.e., with the plane wave as illumination only a single reflected and a single transmitted plane wave emerge. However, this no longer holds when considering metamaterials with nonlocal constitutive relations where we just learned that multiple plane waves can be excited for a given transverse wave vector. To fix the amplitudes of these plane waves, additional interface conditions are required, i.e., interface conditions in addition to those usually considered. This will be done in the following section for the material models of interest.

Prior starting with the derivation, we introduce some notations that will be used in the following sections. This is necessary to lighten to some extent the notations in order to keep it readable. Below, Ω is an *open* domain in $\mathbb{R}^n$.

(1) By $\mathbf{r} = (x,y,z)$ we denote points in $\mathbb{R}^3$ (spatial variable), by $t \in \mathbb{R}$ we denote a time variable.

(2) $\mathbb{R}^3_+ := \{\mathbf{r} \in \mathbb{R}^3 : \ z > 0\}$, $\mathbb{R}^3_- := \{\mathbf{r} \in \mathbb{R}^3 : \ z < 0\}$, $\Gamma = \{\mathbf{r} \in \mathbb{R}^3 : \ z = 0\}$. They denote the different half-spaces above and below the interface we consider in the derivation of the interface conditions.

(3) $\mathbf{n} = (0,0,1)^{\mathrm{T}}$ is the unit normal vector on Γ with $^{\mathrm{T}}$ is the operation of transposition.

(4) For $\Psi : \Gamma \to \mathbb{C}$, we set $\nabla_\Gamma \Psi := (\partial_x \Psi, \partial_y \Psi, 0)^{\mathrm{T}}$.

(5) For $\Phi = (\Phi_x, \Phi_y, \Phi_z) : \Gamma \to \mathbb{C}^3$, we set $\nabla_\Gamma \cdot \Phi := \partial_x \Phi_x + \partial_y \Phi_y$.

(6) $\mathsf{C}^m(\Omega)$, $m \leqslant \infty$, is the space of functions with continuous derivatives up to order m on Ω.

(7) $\mathsf{C}^m(\overline{\Omega})$, $m \leqslant \infty$, is the space of all restrictions of functions in $\mathsf{C}^m(\mathbb{R}^n)$ to $\overline{\Omega}$.

(8) $\mathsf{C}^m_0(\Omega)$ is the space of functions $f \in \mathsf{C}^m(\Omega)$ having compact support in Ω (i.e., $\mathrm{supp}(f) := \overline{\{x : \ f(x) \neq 0\}}$ is a compact set contained in Ω).

(9) $\mathsf{D}'(\Omega)$ is the space of generalized scalar functions on Ω [i.e., linear continuous functionals acting on $\mathsf{C}^\infty_0(\Omega)$].

(11) $\mathsf{L}^2(\Omega)$ is the space of measurable functions $\Psi : \Omega \to \mathbb{C}$ satisfying $\int_\Omega |\Psi(\mathbf{r})|^2 \, d\mathbf{r} < \infty$.

(12) $\mathsf{H}^1(\Omega)$ is the space of measurable functions $\Psi : \Omega \to \mathbb{C}$ such that $\Psi \in \mathsf{L}^2(\Omega)$ and each component of $\nabla\Psi$ belongs to $\mathsf{L}^2(\Omega)$. Hereinafter, differential operations are understood in the generalized sense.

(13) $\mathbf{C}^m(\Omega)$ (resp. $\mathbf{C}^m(\overline{\Omega})$, $\mathbf{C}^m_0(\Omega)$, $\mathbf{L}^2(\Omega)$, $\mathbf{H}^1(\Omega)$) is the space of vector functions $\Phi : \Omega \to \mathbb{C}^3$ with components being in $\mathsf{C}^m(\Omega)$ [respectively, $\mathsf{C}^m(\overline{\Omega})$, $\mathsf{C}^m_0(\Omega)$, $\mathsf{L}^2(\Omega)$, $\mathsf{H}^1(\Omega)$].

(14) $\mathbf{D}'(\Omega)$ is the space of generalized vector functions on Ω [i.e., linear functionals acting on $\mathbf{C}^\infty_0(\Omega)$].

(15) $\mathsf{L}^2_{\mathrm{loc}}(\Omega)$ [respectively, $\mathbf{L}^2_{\mathrm{loc}}(\Omega)$, $\mathsf{H}^1_{\mathrm{loc}}(\Omega)$, $\mathbf{H}^1_{\mathrm{loc}}(\Omega)$] is the space of (scalar or vector-valued) functions belonging to $\mathsf{L}^2(\widehat{\Omega})$ [respectively, $\mathbf{L}^2(\widehat{\Omega})$, $\mathsf{H}^1(\widehat{\Omega})$, $\mathbf{H}^1(\widehat{\Omega})$] for each bounded subdomain $\widehat{\Omega} \subset \Omega$.

(16) I is the identity (3×3) matrix.

We assume that the upper half-space $\mathbb{R}^3_+$ is occupied by vacuum, while the lower subspace $\mathbb{R}^3_-$ is occupied by a homogeneous metamaterial. We want to derive the interface conditions for this situation. Recall that Γ is the interface between them. In the following, we drop the space and frequency (or vacuum wave-number) arguments from our expressions to keep it more light. From the considerations above, we know that we can write Maxwell's equations in terms of a wave-type equation that reads as

$$\nabla \times \nabla \times \mathbf{E} = k_0^2(\mathbf{E} + \mathbf{P}(\mathbf{E})). \tag{34}$$

For the upper half space, $z > 0$, one has $\mathbf{P} \equiv 0$, while for $z < 0$ (i.e., in the metamaterial) we assume the combination of the constitutive relations derived in Sec. II. Please note that our treatment is now on the fourth-order model and the second-order model that retains the terms necessary because of the symmetry of the structure simultaneously for convenience. Application to a specific model can be easily enforced by setting the respective other parameters to zero. The polarization in the metamaterial region, therefore, reads as

$$\mathbf{P}(\mathbf{E}) = (\varepsilon - 1)\mathbf{E} + \nabla \times \alpha(\nabla \times \mathbf{E}) + \sum_{j\in\{x,y,z\}} \partial_j(\beta^j \partial_j \mathbf{E}) + \nabla \times \nabla \times \gamma(\nabla \times \nabla \times \mathbf{E}). \tag{35}$$

Here $\varepsilon,\alpha,\beta^j,\gamma : \overline{\mathbb{R}^3_-} \to \mathbb{C}^{3\times 3}$ are matrix functions with C^∞-smooth and bounded entries. They depend as considered before on the frequency, i.e., the material properties are dispersive, but the frequency dependency is suppressed from now on to simplify the notation. As we consider homogenous metamaterials, they do not depend on the spatial coordinates. Thus the considered wave equation can be rewritten as

$$\frac{\nabla \times \nabla \times \mathbf{E}}{k_0^2} = \tilde{\varepsilon}\mathbf{E} + \nabla \times \tilde{\alpha}(\nabla \times \mathbf{E}) + \sum_{j\in\{x,y,z\}} \partial_j(\tilde{\beta}^j \partial_j \mathbf{E}) + \nabla \times \nabla \times \tilde{\gamma}(\nabla \times \nabla \times \mathbf{E}), \tag{36}$$

where

$$\tilde{\varepsilon} = \begin{cases} \varepsilon, & z<0, \\ 1, & z>0, \end{cases} \quad \tilde{\alpha} = \begin{cases} \alpha, & z<0, \\ 0, & z>0, \end{cases}$$

$$\tilde{\beta}^j = \begin{cases} \beta^j, & z<0, \\ 0, & z>0, \end{cases} \quad \tilde{\gamma} = \begin{cases} \gamma, & z<0, \\ 0, & z>0. \end{cases}$$

V. GENERALISED SOLUTIONS

Up to now our considerations were rather formal. Now it is time to ask: in which sense has Eq. (36) to be understood? Since the matrix functions $\tilde{\varepsilon},\tilde{\alpha},\tilde{\beta}^j,\tilde{\gamma}$ are only piecewise continuous, we are not allowed to regard the differential expression on the right-hand side of Eq. (36) in the classical sense. The natural idea is then to treat this equation in a suitable generalized sense. From these considerations, we can extract already specific requirements concerning the different functions, in particular concerning the spaces in which they are defined. This is helpful in the following sections as interface conditions can be found that require the mere analysis of the space in which these functions are living.

Recall (see, e.g., Ref. [38] for more details) that the notation $\mathbf{D}'(\mathbb{R}^3)$ stands for the space of generalized functions (distributions), i.e., linear functionals acting continuously on $\mathbf{C}_0^\infty(\mathbb{R}^3)$. For example, for $\mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$, $\nabla \times \mathbf{E} \in \mathbf{D}'(\mathbb{R}^3)$ is defined by the action

$$(\nabla \times \mathbf{E})[\Phi] := \int_{\mathbb{R}^3} \mathbf{E} \cdot (\nabla \times \Phi)\, d\mathbf{r}, \ \Phi \in \mathbf{C}_0^\infty(\mathbb{R}^3),$$

which mimics partial integration. Also higher derivatives of $\mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$ can be defined analogously, but the discontinuous function $\tilde{\alpha}$ on the right-hand side of Eq. (36) causes difficulties in mimicking partial integration (which would be no problem if $\tilde{\alpha}$ was a C^1-function). We can solve this difficulty by requiring the additional regularity condition $\nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_-)$. Under this assumption, the product $\tilde{\alpha}(\nabla \times \mathbf{E})$ is a (regular) generalized function, which is defined by $\alpha(\nabla \times \mathbf{E})$ in $\mathbb{R}^3_-$ and 0 in $\mathbb{R}^3_+$ and now we are able to define its generalized $\nabla\times$ derivative. Actually, we require even $\nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$, in order to treat the *left-hand side* of (38) appropriately when defining generalized solutions. Note that the property $\nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$ is not guaranteed by the requirement that $\nabla \times \mathbf{E}$ is in $\mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_+)$ and in $\mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_-)$, but also (some kind of) continuity of the tangential component of $\mathbf{E}$ at the interface is needed.

Analogous considerations for the other terms in Eq. (36) lead to the following natural regularity assumptions:

$$\mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3), \quad \nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3),$$
$$\mathbf{E} \in \mathbf{H}^1_{\mathrm{loc}}(\mathbb{R}^3_-), \quad \nabla \times \nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_-). \tag{37}$$

Now, Eq. (36) is understood as an equality in $\mathbf{D}'(\mathbb{R}^3)$. By the definition of the generalized derivatives, mimicking partial integration in all occurring terms, it is equivalent to

$$\forall \Phi \in \mathbf{C}_0^\infty(\mathbb{R}^3): \int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot (\nabla \times \Phi)\, d\mathbf{r} = k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \Phi\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \Bigg(\varepsilon \mathbf{E} \cdot \Phi + \alpha(\nabla \times \mathbf{E}) \cdot (\nabla \times \Phi) - \sum_{j\in\{x,y,z\}} \beta^j \partial_j \mathbf{E} \cdot \partial_j \Phi + \gamma(\nabla \times \nabla \times \mathbf{E}) \cdot (\nabla \times \nabla \times \Phi) \Bigg) d\mathbf{r}. \tag{38}$$

The vector function $\mathbf{E} : \mathbb{R}^3 \to \mathbb{C}$ is said to be a *generalized* (or *weak*) solution to Eq. (36) if it meets the regularity properties given in conditions (37) and satisfies Eq. (38).

Note that one can introduce another definition of the generalized solution in which the requirement $\nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$ is omitted: we say that $\mathbf{E} : \mathbb{R}^3 \to \mathbb{C}$ is a *very weak solution* to (36) if $\mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$, $\mathbf{E} \in \mathbf{H}^1_{\mathrm{loc}}(\mathbb{R}^3_-)$ (implying also $\nabla \times \mathbf{E} \in \mathbf{L}^2(\mathbb{R}^3_-)$), $\nabla \times \nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_-)$ and $\forall \Phi \in \mathbf{C}_0^\infty(\mathbb{R}^3)$ $\int_{\mathbb{R}^3} \mathbf{E} \cdot (\nabla \times \nabla \times \Phi)\, d\mathbf{r} = \mathrm{RHS}_{(38)}$, where $\mathrm{RHS}_{(38)}$ denotes the right-hand-side of equality (38). Evidently, if $\mathbf{E}$ is a weak solution to (36) then it is also a very weak solution (apparently the opposite is not true). In this paper, we focus on weak solutions since for local constitutive relations (i.e., $\alpha = \gamma = \beta^j = 0$) they satisfy classical interface conditions $[\mathbf{E} \times n]$, $[\tilde{\varepsilon}\mathbf{E} \cdot n]$, $[(\nabla \times \mathbf{E}) \times n]$, $[(\nabla \times \mathbf{E}) \cdot n] = 0$ on Γ (here $[\ldots]$ stands for the jump of the enclosed quantity across Γ)—this follows immediately from our results below.

We also remark that analogous considerations can be performed for $\mathbf{L}^1$-type spaces. Nevertheless, we prefer to deal with $\mathbf{L}^2$ functions and spaces, since we expect that the $\mathbf{L}^2$ setting yields more benefits in subsequent research (in particular, since $\mathbf{L}^2$ is a Hilbert space).

Let $\mathbf{E}$ be a generalized solution to Eq. (36). It is easy to show, just by taking $\Phi \in \mathbf{C}_0^\infty(\mathbb{R}^3_+)$ in Eq. (38), that

$$\nabla \times \nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_+) \tag{39}$$

and

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \mathbf{E} \text{ for almost all } x \in \mathbb{R}^3_+. \tag{40}$$

"Almost all" means that the set consisting of points $x \in \mathbb{R}^3_+$ at which the property expressed in Eq. (40) fails has a Lebesque measure zero.

Similarly, taking $\Phi \in \mathbf{C}_0^\infty(\mathbb{R}^3_-)$, we conclude that

$$\nabla \times \alpha(\nabla \times \mathbf{E}) + \sum_{j\in\{x,y,z\}} \partial_j(\beta^j \partial_j \mathbf{E}) + \nabla \times \nabla \times \gamma(\nabla \times \nabla \times \mathbf{E}) \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3_-) \tag{41}$$

and

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \left(\varepsilon \mathbf{E} + \nabla \times \alpha(\nabla \times \mathbf{E}) + \sum_{j\in\{x,y,z\}} \partial_j(\beta^j \partial_j \mathbf{E}) + \nabla \times \nabla \times \gamma(\nabla \times \nabla \times \mathbf{E}) \right), \tag{42}$$

for almost all $x \in \mathbb{R}^3_-$. Note that the regularity property given in expression (41) means that the sum of the generalized functions that appear in this expression are in $\mathbf{L}^2_{\text{loc}}(\mathbb{R}^3_-)$, but it does not imply that the individual generalized functions belong to $\mathbf{L}^2_{\text{loc}}(\mathbb{R}^3_-)$. Now that we have analyzed generalized solutions outside the interface Γ, we can derive the actual interface conditions in the next section.

VI. DERIVATION OF THE INTERFACE CONDITIONS

Let $\mathbf{E}$ be a generalized solution to Eq. (36). We denote by $\mathbf{E}_+$ and $\mathbf{E}_-$ the restrictions of $\mathbf{E}$ to $\mathbb{R}^3_+$ and $\mathbb{R}^3_-$, respectively. Also, we additionally assume in this section that $\mathbf{E}$ is smooth in each half-space, namely,

$$\mathbf{E}_+ \in \mathsf{C}^4(\overline{\mathbb{R}^3_+}),\ \mathbf{E}_- \in \mathsf{C}^4(\overline{\mathbb{R}^3_-}). \tag{43}$$

These additional smoothness conditions are indeed satisfied if the coefficients α, β^j, γ are smooth, at least in the case where the differential Eq. (36) is elliptic (e.g., if γ is positive definite); see Ref. [39]. Note, that the interface conditions we are going to derive in this section under the additional smoothness assumption expressed in (43) also remain valid when only our conditions (37) are satisfied. However, then they hold only in some generalized sense which needs the concept of traces. Being aware that the discussion on traces and generalized interface conditions is rather of interest for mathematical audience, we do not include these issues into the manuscript. Nevertheless, one can find them in our preprint [40], which also contains an appendix with a short introduction to the theory of generalized functions.

Below, in volume integrals we will use the notation $\mathbf{E}$, while in integrals over Γ we deal with $\mathbf{E}_+$ and $\mathbf{E}_-$. In the following, we distinguish between the main interface conditions and two alternative interface conditions. These alternative interface conditions do not contain any further information and indeed can be derived from the main interface conditions. However, their documentation seems useful as these alternative interface conditions look simpler. This may make them more suitable for use in some specific situations.

A. Main interface conditions

In the following, we want to prove that if $\mathbf{E}$ is a generalized solution of Eq. (36) and satisfies the regularity assumptions (43), then $\mathbf{E}$ satisfies the following interface conditions on Γ:

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0, \tag{C$_1$}$$

$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} - k_0^2(\mathrm{I} - \mathbf{n}\mathbf{n}^{\mathrm{T}})\beta^z \partial_z \mathbf{E}_- + k_0^2(\alpha \nabla \times \mathbf{E}_- + \nabla \times \gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0, \tag{C$_2$}$$

$$(\gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0, \tag{C$_3$}$$

$$(\beta^z \partial_z \mathbf{E}_-) \cdot \mathbf{n} = 0 \tag{C$_4$}$$

Conversely, if $\mathbf{E}$ satisfies (43), solves (40) in $\mathbb{R}^3_+$, solves (42) in $\mathbb{R}^3_-$ and the conditions (C$_1$)–(C$_4$) are fulfilled, then $\mathbf{E}$ is a generalized solution to (36). Please note, these interface conditions are one of the central results from our contributions. In the following, we prove each of them.

1. Proof of the first interface condition

Since $\mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3)$ and $\nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3)$, we have $\forall \Phi \in \mathbf{C}_0^\infty(\mathbb{R}^3)$:

$$\int_{\mathbb{R}^3} \mathbf{E} \cdot (\nabla \times \Phi)\, d\mathbf{r} = \int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot \Phi\, d\mathbf{r}. \tag{44}$$

On the other hand, by integrating by parts in each half-space, we obtain

$$\begin{aligned}\int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot \Phi\, d\mathbf{r} &= \int_{\mathbb{R}^3_-} (\nabla \times \mathbf{E}) \cdot \Phi\, d\mathbf{r} + \int_{\mathbb{R}^3_+} (\nabla \times \mathbf{E}) \cdot \Phi\, d\mathbf{r} \\ &= \int_{\mathbb{R}^3_-} \mathbf{E} \cdot (\nabla \times \Phi)\, d\mathbf{r} + \int_{\mathbb{R}^3_+} \mathbf{E} \cdot (\nabla \times \Phi)\, d\mathbf{r} + \int_\Gamma (\mathbf{E}_+ \times \mathbf{n} - \mathbf{E}_- \times \mathbf{n}) \cdot \Phi\, ds \\ &= \int_{\mathbb{R}^3} \mathbf{E} \cdot (\nabla \times \Phi)\, d\mathbf{r} + \int_\Gamma (\mathbf{E}_+ \times \mathbf{n} - \mathbf{E}_- \times \mathbf{n}) \cdot \Phi\, ds,\end{aligned} \tag{45}$$

where $d\mathbf{s} = dx\, dy$ is the area of the surface element on Γ. Since Φ is arbitrary, we obtain from Eqs. (44) and (45) the first interface conditions (C_1).

2. Proof of the second interface condition

To prove the second interface condition, we decompose each integral in Eq. (38) in a sum of integrals over $\mathbb{R}^3_-$ and $\mathbb{R}^3_+$ and integrate by parts in such a way that all the derivatives shift from Φ to $\mathbf{E}$. Then, moving all volume integrals to the left-hand side and all integrals over Γ to the right-hand side, we get

$$\begin{aligned}
&\int_{\mathbb{R}^3}(\nabla\times\nabla\times\mathbf{E})\cdot\Phi\, d\mathbf{r} - k_0^2\int_{\mathbb{R}^3_+}\mathbf{E}\cdot\Phi\, d\mathbf{r}\\
&\quad -k_0^2\int_{\mathbb{R}^3_-}\left(\varepsilon\mathbf{E}+\nabla\times\alpha\nabla\times\mathbf{E}+\sum_{j\in\{x,y,z\}}\partial_j\big(\beta^j\partial_j\mathbf{E}\big)+\nabla\times\nabla\times\gamma(\nabla\times\nabla\times\mathbf{E}_-)\right)\cdot\Phi\, d\mathbf{r}\\
&= \int_\Gamma \big(\big(\nabla\times\mathbf{E}_+ - \nabla\times\mathbf{E}_- + k_0^2(\alpha\nabla\times\mathbf{E}_- + \nabla\times\gamma\nabla\times\nabla\times\mathbf{E}_-)\big)\times\mathbf{n}\big)\cdot\Phi\, d\mathbf{s}\\
&\quad +k_0^2\int_\Gamma((\gamma\nabla\times\nabla\times\mathbf{E}_-)\times\mathbf{n})\cdot(\nabla\times\Phi)\, d\mathbf{s} - k_0^2\int_\Gamma(\beta^z\partial_z\mathbf{E}_-)\cdot\Phi\, d\mathbf{s}.
\end{aligned}\tag{46}$$

Due to Eqs. (40) and (42), the left-hand side of Eq. (46) vanishes and thus Eq. (46) can be rewritten as follows:

$$\begin{aligned}
&\int_\Gamma \big(\big(\nabla\times\mathbf{E}_+ - \nabla\times\mathbf{E}_- + k_0^2(\alpha\nabla\times\mathbf{E}_- + \nabla\times\gamma\nabla\times\nabla\times\mathbf{E}_-)\big)\times\mathbf{n} - k_0^2(\mathrm{I}-\mathbf{n}\mathbf{n}^{\mathrm{T}})\beta^z\partial_z\mathbf{E}_-\big)\cdot\Phi\, d\mathbf{s}\\
&\quad + k_0^2\int_\Gamma((\gamma\nabla\times\nabla\times\mathbf{E}_-)\times\mathbf{n})\cdot(\nabla\times\Phi)\, d\mathbf{s} - k_0^2\int_\Gamma(\mathbf{n}\mathbf{n}^{\mathrm{T}}\beta^z\partial_z\mathbf{E}_-)\cdot\Phi\, d\mathbf{s} = 0.
\end{aligned}\tag{47}$$

Now, we choose the function Φ of the form

$$\Phi(\mathbf{r}) = (\Phi_1(x,y)\eta(z),\ \Phi_2(x,y)\eta(z),\ 0)^{\mathrm{T}},\tag{48}$$

where $\Phi_1,\Phi_2\in C_0^\infty(\mathbb{R}^2)$, $\eta\in C_0^\infty(\mathbb{R})$, moreover, $\eta|_{\{|z|<\delta\}}=1$ for some $\delta>0$. One has $\nabla\times\Phi = (-\Phi_2\eta', \Phi_1\eta', (\partial_x\Phi_2-\partial_y\Phi_1)\eta)^{\mathrm{T}}$, whence

$$\nabla\times\Phi|_\Gamma = (0,\ 0,\ \partial_x\Phi_2-\partial_y\Phi_1)^{\mathrm{T}},$$

and, hence, the second integral in Eq. (47) vanishes. Since the z component of Φ is equal to zero, the third integral in Eq. (47) also vanishes. Thus

$$\int_\Gamma \big(\big(\nabla\times\mathbf{E}_+ - \nabla\times\mathbf{E}_- + k_0^2(\alpha\nabla\times\mathbf{E}_- + \nabla\times\gamma\nabla\times\nabla\times\mathbf{E}_-)\big)\times\mathbf{n} - k_0^2(\mathrm{I}-\mathbf{n}\mathbf{n}^{\mathrm{T}})\beta^z\partial_z\mathbf{E}_-\big)\cdot(\Phi_1,\Phi_2,0)^{\mathrm{T}}\, d\mathbf{s} = 0.\tag{49}$$

Finally, since the functions Φ_1 and Φ_2 are arbitrary, we conclude from Eq. (49) the interface condition (C_2).

3. Proof of the third interface condition

To prove the third interface condition, we take the function Φ of the form

$$\Phi(\mathbf{r}) = (\Phi_2(x,y)z\eta(z),\ -\Phi_1(x,y)z\eta(z),\ 0)^{\mathrm{T}}.\tag{50}$$

As before, both $\Phi_1,\Phi_2\in C_0^\infty(\mathbb{R}^2)$ and $\eta\in C_0^\infty(\mathbb{R})$, with $\eta|_{\{|z|<\delta\}}=1$ for some $\delta>0$. Then, we get $\nabla\times\Phi = ((\eta+z\eta')\Phi_1, (\eta+z\eta')\Phi_2, (-\partial_x\Phi_1-\partial_y\Phi_2)z\eta)^{\mathrm{T}}$ and hence

$$\Phi|_\Gamma = 0,\quad \nabla\times\Phi|_\Gamma = (\Phi_1,\ \Phi_2,\ 0)^{\mathrm{T}}.\tag{51}$$

We substitute this function Φ into Eq. (47). Due to (51), all integrals in Eq. (47) vanish except the second one. Thus one gets

$$\int_{\Gamma}((\gamma\nabla\times\nabla\times\mathbf{E}_-)\times\mathbf{n})\cdot(\Phi_1,\Phi_2,0)^{\mathrm{T}}\,d\mathbf{s} = 0. \tag{52}$$

Since Φ_1 and Φ_2 are arbitrary, Eq. (52) implies the third interface condition (C_3).

4. Proof of the fourth interface condition

Finally, due to (C_2) and (C_3), the first two integrals in Eq. (47) vanish for any arbitrary function Φ, whence one gets

$$\int_{\Gamma}(\mathbf{n}\mathbf{n}^{\mathrm{T}}\beta^z\partial_z\mathbf{E}_-)\cdot\Phi\,d\mathbf{s} = 0, \tag{53}$$

and, consequently, we arrive at the last condition (C_4).

5. Further remarks

Conversely, let now $\mathbf{E}$ satisfy requirement (43), solve Eq. (40) in $\mathbb{R}^3_+$, solve Eq. (42) in $\mathbb{R}^3_-$, and let (C_1)–(C_4) hold. We have to show that $\mathbf{E}$ is a generalized solution to Eq. (36).

Indeed, it follows from the assumption (43) that $\mathbf{E}\in\mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$, $\mathbf{E}\in\mathbf{H}^1_{\mathrm{loc}}(\mathbb{R}^3_-)$, and $\nabla\times\nabla\times\mathbf{E}\in\mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_-)$. Moreover, due to ($C_1$), $\nabla\times\mathbf{E}\in\mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$. Thus conditions (37) are satisfied. Integrating by parts we get for an arbitrary $\Phi\in\mathbf{C}_0^\infty(\mathbb{R}^3)$:

$$\begin{aligned}-k_0^2\int_{\mathbb{R}^3_-}&\left(\varepsilon\mathbf{E}\cdot\Phi+\alpha(\nabla\times\mathbf{E})\cdot(\nabla\times\Phi)-\sum_{j\in\{x,y,z\}}\beta^j\partial_j\mathbf{E}\cdot\partial_j\Phi+\gamma(\nabla\times\nabla\times\mathbf{E})\cdot(\nabla\times\nabla\times\Phi)\right)d\mathbf{r}\\&+\int_{\mathbb{R}^3}(\nabla\times\mathbf{E})\cdot(\nabla\times\Phi)\,d\mathbf{r}-k_0^2\int_{\mathbb{R}^3_+}\mathbf{E}\cdot\Phi\,d\mathbf{r}=\mathrm{LHS}_{(46)}-\mathrm{RHS}_{(46)},\end{aligned} \tag{54}$$

where $\mathrm{LHS}_{(46)}$ [respectively, $\mathrm{RHS}_{(46)}$] is the expression standing in the left-hand side (respectively, right-hand side) of Eq. (46). Due to (40)–(42), $\mathrm{LHS}_{(46)}=0$ and, due to conditions (C_2)–(C_4), $\mathrm{RHS}_{(46)}=0$. Therefore Eq. (38) holds true and, consequently, $\mathbf{E}$ is a generalized solution to Eq. (36).

With that we have been offering proofs for the main interface conditions. In the following, we formulate two alternative interface conditions that can be used as well, but they are not fundamental since they follow from the definition of the generalized solution and the main interface conditions.

B. Alternative interface conditions

Let $\mathbf{E}$ be a generalized solution to Eq. (36) satisfying the regularity assumptions (43). Then $\mathbf{E}$ meets the interface conditions (C_1)–(C_4). The fulfillment of (C_1)–(C_4) is also a sufficient condition for being a generalized solution if (40) and (42) hold—see the statement after conditions (C_1)–(C_4). In this section, we derive two alternative interface conditions on Γ. They appear slightly simpler and are, therefore, of practical use in further research.

1. First alternative interface condition

Let $\Psi\in\mathrm{C}_0^\infty(\mathbb{R}^3)$ be an arbitrary function. Since $\nabla\times\mathbf{E}\in\mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$, $\nabla\cdot(\nabla\times\mathbf{E})=0$ and $\nabla\times(\nabla\Psi)=0$, one has

$$\begin{aligned}0&=-\int_{\mathbb{R}^3}(\nabla\cdot(\nabla\times\mathbf{E}))\Psi\,d\mathbf{r}=-\int_{\mathbb{R}^3_+}(\nabla\cdot(\nabla\times\mathbf{E}))\Psi\,d\mathbf{r}-\int_{\mathbb{R}^3_-}(\nabla\cdot(\nabla\times\mathbf{E}))\Psi\,d\mathbf{r}\\&=\int_{\Gamma}((\nabla\times\mathbf{E}_+-\nabla\times\mathbf{E}_-)\cdot\mathbf{n})\Psi\,d\mathbf{s}+\int_{\mathbb{R}^3_+}(\nabla\times\mathbf{E})\cdot(\nabla\Psi)\,d\mathbf{r}+\int_{\mathbb{R}^3_-}(\nabla\times\mathbf{E})\cdot(\nabla\Psi)\,d\mathbf{r}\\&=\int_{\Gamma}((\nabla\times\mathbf{E}_+-\nabla\times\mathbf{E}_-)\cdot\mathbf{n})\Psi\,d\mathbf{s}+\int_{\Gamma}((\mathbf{E}_+-\mathbf{E}_-)\times\mathbf{n})\cdot\nabla\Psi\,d\mathbf{s}+\int_{\mathbb{R}^3}\mathbf{E}\cdot(\nabla\times(\nabla\Psi))\,d\mathbf{r}\\&\overset{(C_1)}{=}\int_{\Gamma}((\nabla\times\mathbf{E}_+-\nabla\times\mathbf{E}_-)\cdot\mathbf{n})\Psi\,d\mathbf{s},\end{aligned}$$

whence we get the following interface condition on Γ:

$$(\nabla\times\mathbf{E}_+-\nabla\times\mathbf{E}_-)\cdot\mathbf{n}=0. \tag{C_5}$$

2. Second alternative interface condition

For materials governed by local constitutive relations (i.e., $\alpha = \gamma = \beta^j = 0$) one has also the interface condition

$$(\mathbf{E}_+ - \varepsilon \mathbf{E}_-) \cdot \mathbf{n} = 0, \tag{55}$$

which follows from the fact that $\tilde{\varepsilon}\mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$ and, due to Eq. (36), $\nabla \cdot (\tilde{\varepsilon}\mathbf{E}) = 0$.

Let us now derive an analog of the interface condition in Eq. (55) for our nonlocal model. For this purpose, we substitute into Eq. (38) the function Φ of the form

$$\Phi = \nabla \Psi,$$

where Ψ is a smooth compactly supported scalar function. Under this choice $\nabla \times \Phi = 0$ and Eq. (38) becomes

$$\int_{\mathbb{R}^3_-} \left(\varepsilon \mathbf{E} \cdot \nabla \Psi - \sum_{j \in \{x,y,z\}} \beta^j \partial_j \mathbf{E} \cdot \partial_j \nabla \Psi \right) d\mathbf{r} + \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \nabla \Psi \, d\mathbf{r} = 0. \tag{56}$$

Let us additionally assume that supp$\Psi \subset \mathbb{R}^3_-$; integrating by part in Eq. (56), we get $\forall \Psi \in \mathsf{C}^\infty_0(\mathbb{R}^3_-)$:

$$\int_{\mathbb{R}^3_-} \left(\nabla \cdot \left(\varepsilon \mathbf{E} + \sum_{j \in \{x,y,z\}} \partial_j (\beta^j \partial_j \mathbf{E}) \right) \right) \Psi \, d\mathbf{r} = 0,$$

whence, since $\mathsf{C}^\infty_0(\mathbb{R}^3_-)$ is dense in $\mathsf{L}^2(\mathbb{R}^3_-)$,

$$\nabla \cdot \left(\varepsilon \mathbf{E} + \sum_{j \in \{x,y,z\}} \partial_j (\beta^j \partial_j \mathbf{E}) \right) = 0 \ \text{ in } \mathbb{R}^3_-. \tag{57}$$

Similarly, by using $\Psi \in \mathsf{C}^\infty_0(\mathbb{R}^3_+)$, we obtain

$$\nabla \cdot \mathbf{E} = 0 \ \text{ in } \mathbb{R}^3_+. \tag{58}$$

Now, we take an arbitrary $\Psi \in \mathsf{C}^\infty_0(\mathbb{R}^3)$ in Eq. (56) and integrate by parts in each half-space $\mathbb{R}^3_\pm$. We get

$$\int_\Gamma \left(\left(\mathbf{E}_+ - \varepsilon \mathbf{E}_- - \sum_{j \in \{x,y,z\}} \partial_j (\beta^j \partial_j \mathbf{E}_-) \right) \cdot \mathbf{n} \right) \Psi \, d\mathbf{s} + \int_\Gamma \beta^z \partial_z \mathbf{E}_- \cdot \nabla \Psi \, d\mathbf{s} + \int_{\mathbb{R}^3_+} (\nabla \cdot \mathbf{E}) \, \Psi \, d\mathbf{r}$$
$$+ \int_{\mathbb{R}^3_-} \nabla \cdot \left(\varepsilon \mathbf{E} + \sum_{j \in \{x,y,z\}} \partial_j (\beta^j \partial_j \mathbf{E}) \right) \Psi \, d\mathbf{r} = 0. \tag{59}$$

In this equality, the integrals over $\mathbb{R}^3_\pm$ vanish due to Eq. (57) and Eq. (58). Moreover, in view of interface condition (C_4), the last integral in Eq. (59) can be rewritten as follows:

$$\int_\Gamma \beta^z \partial_z \mathbf{E}_- \cdot \nabla \Psi \, d\mathbf{s} = \int_\Gamma (\mathrm{I} - \mathbf{n}\mathbf{n}^{\mathrm{T}}) \beta^z \partial_z \mathbf{E}_- \cdot \nabla_\Gamma \Psi \, d\mathbf{s} = - \int_\Gamma \nabla_\Gamma \cdot (\beta^z \partial_z \mathbf{E}_-) \, \Psi \, d\mathbf{s}. \tag{60}$$

Since $\Psi \in \mathsf{C}^\infty_0(\mathbb{R}^3)$ is an arbitrary function, we conclude from Eqs. (59)–(60) the following interface condition on Γ:

$$\left(\mathbf{E}_+ - \varepsilon \mathbf{E}_- - \sum_{j \in \{x,y,z\}} \partial_j (\beta^j \partial_j \mathbf{E}_-) \right) \cdot \mathbf{n} - \nabla_\Gamma \cdot (\beta^z \partial_z \mathbf{E}_z) = 0. \tag{C$_6$}$$

That closes this section. We will use these interface conditions to derive Fresnel equations for the transmission and reflection of a plane wave from a slab in the next section.

VII. FRESNEL FORMULAS

In this section, we apply the interface conditions obtained above to the problem of light propagation through a slab of metamaterial. The geometry of the pertinent problem hence is defined as $\Omega = \{\mathbf{r} \in \mathbb{R}^3 : -\delta < z < 0\}$, $\Omega_- = \{\mathbf{r} \in \mathbb{R}^3 : z < -\delta\}$, $\Omega_+ = \{\mathbf{r} \in \mathbb{R}^3 : z > 0\}$, where $\delta > 0$. We assume that Ω is filled with a metamaterial, which is governed by the constitutive relations expressed in Eq. (35). Moreover, additionally, we assume that the coordinate system of the laboratory corresponds to the coordinate system of the principle axis of the metamaterial. The materials considered here are centrosymmetric. Therefore the material tensors are considered to be diagonal. In essence, this means that the unit cells are aligned to the slab we consider. Note that this is not an essential condition but it helps us to keep the expressions sufficiently simple. The remaining space (that is $\Omega_- \cup \Omega_+$) is occupied by vacuum.

We also denote

$$\Gamma_- = \{\mathbf{r} \in \partial\Omega : \ z = -\delta\}, \quad \Gamma_+ = \{\mathbf{r} \in \partial\Omega : \ z = 0\}.$$

Now, assume that we have an incident plane wave impinging on the slab from Ω_+. A part of this wave will be reflected, the other part will be transmitted through the slab to Ω_-. Our goal is to find the amplitudes of these reflected and transmitted waves, in other words, we want to derive Fresnel-type formulas. Even though discussed here for a plane wave, an arbitrary illumination can always be written as a superposition of plane waves. Therefore the plane-wave assumption is by no means a limitation.

We notice that, due to the special form of the material coefficients expressed in Eqs. (13) and (24), each solution $\mathbf{E}$ of Eq. (36) can be represented in the form

$$\mathbf{E} = \mathbf{E}^{\mathrm{TE}} + \mathbf{E}^{\mathrm{TM}},$$

where $\mathbf{E}^{\mathrm{TE}} = (E_x^{\mathrm{TE}},0,0)^{\mathrm{T}}$ (transverse-electric-polarized wave) and $\mathbf{E}^{\mathrm{TM}} = (0,E_y^{\mathrm{TM}},E_z^{\mathrm{TM}})^{\mathrm{T}}$ (transverse-magnetic-polarized wave), each of which satisfies Eq. (36). In what follows, we treat TE- and TM-polarized incident waves separately. In this section, $\mathbf{r}$ is treated as vector column, i.e., $\mathbf{r} = (x,y,z)^{\mathrm{T}}$.

A. TE polarization

Assume that

$$\mathbf{E}^I = \mathbf{E}_0^I \exp(\mathrm{i}\mathbf{k}^I \cdot \mathbf{r})$$

is the incident TE-polarized plane wave. Here, $\mathbf{k}^I = (k_x^I,k_y^I,k_z^I)^{\mathrm{T}}$ is the wave vector and $\mathbf{E}_0^I = (E_x^I,0,0)^{\mathrm{T}}$ is the amplitude vector of the incident plane wave. The wave vector in vacuum obeys the ordinary dispersion relation, i.e., $k^2 = k_0^2 = \frac{\omega^2}{c^2}$. Moreover, in view of (58) one has $k_x^I = 0$.

Due to symmetry arguments, it is reasonable to search the reflected and the transmitted fields in the same form as the incident field. Namely, the reflected field is searched in the form $\mathbf{E}^R = \mathbf{E}_0^R \exp(\mathrm{i}\mathbf{k}^R \cdot \mathbf{r})$, where $\mathbf{k}^R = (0,k_y^R,k_z^R)^{\mathrm{T}}$, $\mathbf{E}_0^R = (E_x^R,0,0)^{\mathrm{T}}$. The total field in Ω_+ is $\mathbf{E}^I + \mathbf{E}^R$. In Ω_-, we have the transmitted field $\mathbf{E}^T = \mathbf{E}_0^T \exp(\mathrm{i}\mathbf{k}^T \cdot \mathbf{r})$, where $\mathbf{k}^T = (0,k_y^T,k_z^T)^{\mathrm{T}}$, $\mathbf{E}_0^T = (E_x^T,0,0)^{\mathrm{T}}$.

Finally, in the slab Ω, the total field has the form $\mathbf{E}^{\mathrm{slab}} = \sum_{j=1}^N \mathbf{E}_0^j \exp(\mathrm{i}\mathbf{k}^j \cdot \mathbf{r})$, where $\mathbf{k}^j = (0,k_y^j,k_z^j)^{\mathrm{T}}$, $\mathbf{E}_0^j = (E_x^j,0,0)^{\mathrm{T}}$, where N is the number of linearly independent eigenmodes existing in Ω. The larger number of plane waves is reminiscent to the fact that for each value of k_x and k_y multiple solutions for k_z exist at each frequency. This will be discussed below. Also, the field inside the slab is always written as a superposition of forward and backward propagating modes in the principle propagation direction. Therefore this quite general *Ansatz* is chosen.

Plugging the plane-wave *Ansätze*, for example, into (C_1) on Γ_+ and Γ_-, we get the equations

$$E^I \mathrm{e}^{\mathrm{i}(k_x^I x + k_y^I y)} + E^R \mathrm{e}^{\mathrm{i}(k_x^R x + k_y^R y)} - \sum_{j=1}^N E_x^j \mathrm{e}^{\mathrm{i}(k_x^j x + k_y^j y)} = 0,$$

$$\sum_{j=1}^N E_x^j \mathrm{e}^{-\mathrm{i}k_z^j\delta} \mathrm{e}^{\mathrm{i}(k_x^j x + k_y^j y)} - E^T \mathrm{e}^{-\mathrm{i}k_z^T\delta} \mathrm{e}^{\mathrm{i}(k_x^T x + k_y^T y)} = 0,$$

which hold for all $(x,y) \in \mathbb{R}^2$. As the system is translational invariant along the interface, we require all plane waves involved in the process to share the same wave-vector components tangential to the interface. Hence the vectors $\mathbf{k}^I$, $\mathbf{k}^R$, $\mathbf{k}^T$ and $\mathbf{k}^j$ $(j = 1,\ldots,N)$ have the same y components. Hereafter, for the y component of all wave vectors, we use the notation k_y.

It is easy to see that $N = 4$ provided $\gamma_x \neq 0$. Indeed, substituting $\mathbf{E} = (E_x,0,0)^{\mathrm{T}} \exp(\mathrm{i}\mathbf{k} \cdot \mathbf{r})$ (with $k_x = 0$) into Eq. (34) supplemented with the constitutive relations expressed in Eq. (35), we arrive at the following dispersion relation linking k_0 and $\mathbf{k}$ for the metamaterial:

$$k_y^2 + k_z^2 = k_0^2\big(\varepsilon_x + \big(\alpha_z k_y^2 + \alpha_y k_z^2\big) + \gamma_x\big(k_y^2 + k_z^2\big)^2\big). \quad (61)$$

This is a fourth-order polynomial equation with respect to k_z, thus generically we get four eigenmodes.[1] They come in pairs and two of these eigenmodes are forward and two of these eigenmodes are backward propagating. Please note that in this section the propagation goes to the negative z direction, such that the negative branches of k_z has to be chosen.

There are six unknowns E_x^R, E_x^T, E_x^j $(j = 1,\ldots,4)$. On each interface Γ_+ and Γ_- we can initially impose four conditions (C_1)–(C_4), but (C_4) simply reads $0 = 0$. As a result, we have three nontrivial equations on each interface. This leaves us with a total number of six linearly independent equations which is just enough to solve uniquely for all involved amplitudes.

Plugging our plane-wave *Ansätze* into these equations, we arrive at the following linear algebraic system for $E = (E_x^R,E_x^1,E_x^2,E_x^3,E_x^4,E_x^T)^{\mathrm{T}}$:

$$\mathcal{A}E = \mathcal{F},$$

where

$$\mathcal{A} = \begin{pmatrix} 1 & -1 & -1 & -1 & -1 & 0 \\ k_z^R & -A_1 & -A_2 & -A_3 & -A_4 & 0 \\ 0 & \gamma_x(\mathbf{k}^1)^2 & \gamma_x(\mathbf{k}^2)^2 & \gamma_x(\mathbf{k}^3)^2 & \gamma_x(\mathbf{k}^4)^2 & 0 \\ 0 & -\mathrm{e}^{-\mathrm{i}k_z^1\delta} & -\mathrm{e}^{-\mathrm{i}k_z^2\delta} & -\mathrm{e}^{-\mathrm{i}k_z^3\delta} & -\mathrm{e}^{-\mathrm{i}k_z^4\delta} & \mathrm{e}^{-\mathrm{i}k_z^T\delta} \\ 0 & -A_1\mathrm{e}^{-\mathrm{i}k_z^1\delta} & -A_2\mathrm{e}^{-\mathrm{i}k_z^2\delta} & -A_3\mathrm{e}^{-\mathrm{i}k_z^3\delta} & -A_4\mathrm{e}^{-\mathrm{i}k_z^4\delta} & k_z^T\mathrm{e}^{-\mathrm{i}k_z^T\delta} \\ 0 & \gamma_x(\mathbf{k}^1)^2\mathrm{e}^{-\mathrm{i}k_z^1\delta} & \gamma_x(\mathbf{k}^2)^2\mathrm{e}^{-\mathrm{i}k_z^2\delta} & \gamma_x(\mathbf{k}^3)^2\mathrm{e}^{-\mathrm{i}k_z^3\delta} & \gamma_x(\mathbf{k}^4)^2\mathrm{e}^{-\mathrm{i}k_z^4\delta} & 0 \end{pmatrix}, \quad \mathcal{F} = -E_x^I\begin{pmatrix} 1 \\ k_z^I \\ 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}.$$

[1] We omit the special case, when Eq. (61) has multiple roots. This case requires a separate treatment.

Here, $(\mathbf{k}^j)^2 := (k_y)^2 + (k_z^j)^2$, and $A_j := k_z^j(1 - k_0^2(\alpha_y + \gamma_x(\mathbf{k}^j)^2))$. Thus the required amplitudes E_x^R and E_x^T are determined by

$$E^R = [(\mathcal{A}^{-1}\mathcal{F})]_1, \quad E^T = [(\mathcal{A}^{-1}\mathcal{F})]_6,$$

where $[\cdot]_k$ denotes the kth component of a vector. It is not hard to show that generically the matrix $\mathcal{A}$ is invertible; "generically" means that the set consisting of $\varepsilon,\alpha,\gamma,\beta^j$ for which $\det(\mathcal{A}) = 0$ has a Lebesgue measure zero in the space of all admissible parameters. Note that the equation coming from conditions (C_5) coincides with the equation coming from condition (C_1) multiplied by k_y, and the equation coming from condition (C_6) reads $0 = 0$.

B. TM polarization

Again, in Ω_+ the total field is of the form $\mathbf{E}^I + \mathbf{E}^R$, where $\mathbf{E}^I = \mathbf{E}_0^I \exp(\mathrm{i}\mathbf{k}^I \cdot \mathbf{r})$ is the incident field, $\mathbf{E}^R = \mathbf{E}_0^R \exp(\mathrm{i}\mathbf{k}^R \cdot \mathbf{r})$ is the reflected field, but now the fields are TM-polarized, i.e., $\mathbf{E}_0^I = (0, E_y^I, E_z^I)^{\mathrm{T}}, \mathbf{E}_0^R = (0, E_y^R, E_z^R)^{\mathrm{T}}$. In Ω_-, we get the transmitted field $\mathbf{E}^T = \mathbf{E}_0^T \exp(\mathrm{i}\mathbf{k}^T \cdot \mathbf{r})$, where $\mathbf{E}_0^T = (0, E_y^T, E_z^T)^{\mathrm{T}}$. Finally, in the slab Ω, the total field has the form $\mathbf{E}^{\text{slab}} = \sum_{j=1}^{N} \mathbf{E}_0^j \exp(\mathrm{i}\mathbf{k}^j \cdot \mathbf{r})$, where $\mathbf{E}_0^j = (0, E_y^j, E_z^j)^{\mathrm{T}}$.

Again, the vectors $\mathbf{k}^I$, $\mathbf{k}^R$, $\mathbf{k}^T$, and $\mathbf{k}^j$ $(j = 1, \ldots, N)$ have the same x and y components. Similarly to the TE polarization, we denote by k_y the y component of the wave vectors, the x components are chosen to be equal to zero.

For TM polarization, the dispersion relation reads

$$\det\begin{pmatrix} -k_0^2\varepsilon_y + k_z^2 Q + k_0^2\beta_y k_y^2 & -k_y k_z Q \\ -k_y k_z Q & -k_0^2\varepsilon_z + k_y^2 Q + k_0^2\beta_z k_z^2 \end{pmatrix} = 0, \tag{62}$$

where $Q := 1 - k_0^2(\alpha_x + \gamma_z k_y^2 + \gamma_y k_z^2)$. It is easy to see that this is a 6^{th}-order polynomial equation with respect to k_z provided $\gamma_y \neq 0$ and $\beta_z \neq 0$. Thus $N = 6$. There are three forward and three backward propagating modes.

In $\Omega_+ \cup \Omega_-$, one has $\operatorname{div} \mathbf{E} = 0$ and consequently

$$E_y k_y + E_z k_z = 0. \tag{63}$$

In Ω, one has $\operatorname{div}(\varepsilon\mathbf{E} + \sum_{j\in\{x,y,z\}} \partial_j(\beta^j \partial_j \mathbf{E})) = 0$ and consequently

$$E_y k_y (\varepsilon_y - \beta_y k_y^2) + E_z k_z (\varepsilon_z - \beta_z k_z^2) = 0. \tag{64}$$

Due to Eqs. (63) and (64), it is enough to determine only the z components of the reflected and transmitted fields [except in the special case $(\varepsilon_y - \beta_y k_y^2) k_y = 0$, which we do not treat here].

We have eight unknowns E_z^R, E_z^T, E_z^j $(j = 1, \ldots, 6)$. On each interface Γ_+ and Γ_-, we have four conditions (C_1)–(C_4), thus, both interfaces produce totally eight equations. Plugging the plane-wave *Ansätze* into these equations, we arrive at the following system for $E = (E_z^R, E_z^1, E_z^2, E_z^3, E_z^4, E_z^5, E_z^6, E_z^T)^{\mathrm{T}}$: $\mathcal{A}E = \mathcal{F}$, where

$$\mathcal{A} = \left(\begin{array}{c|c|c|c|c|c|c|c} & j=1 & j=2 & j=3 & j=4 & j=5 & j=6 & \\ k_z^R & -B_1 k_z^1 & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ (\mathbf{k}^R)^2 & \left(-1 + k_0^2\alpha_x + k_0^2\left(k_y^2\gamma_z + \left(k_z^1\right)^2\gamma_y\right)\right)\left(k_y^2 + \left(k_z^1\right)^2 B_1\right) & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ 0 & \gamma_y k_z^1\left(k_y^2 + \left(k_z^1\right)^2 B_1\right) & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ 0 & \beta_z k_z^1 & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ 0 & -B_1 k_z^1 \mathrm{e}^{-\mathrm{i}k_z^1\delta} & \cdots & \cdots & \cdots & \cdots & \cdots & k_z^T \mathrm{e}^{-\mathrm{i}k_z^T\delta} \\ 0 & \left(-1 + k_0^2\alpha_x + k_0^2\left(k_y^2\gamma_z + \left(k_z^1\right)^2\gamma_y\right)\right)\left(k_y^2 + (k_z^1)^2 B_1\right)\mathrm{e}^{-\mathrm{i}k_z^1\delta} & \cdots & \cdots & \cdots & \cdots & \cdots & (\mathbf{k}^T)^2 \mathrm{e}^{-\mathrm{i}k_z^T\delta} \\ 0 & \gamma_y k_z^1\left(k_y^2 + \left(k_z^1\right)^2 B_1\right)\mathrm{e}^{-\mathrm{i}k_z^1\delta} & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \\ 0 & \beta_z k_z^1 \mathrm{e}^{-\mathrm{i}k_z^1\delta} & \cdots & \cdots & \cdots & \cdots & \cdots & 0 \end{array}\right),$$

$$\mathcal{F} = -E_z^I\left(k_z^I,\ (\mathbf{k}^I)^2,\ 0,\ 0,\ 0,\ 0,\ 0,\ 0\right)^{\mathrm{T}},$$

and $B_j := \frac{\varepsilon_z - (k_z^j)^2\beta_z}{\varepsilon_y - (k_y)^2\beta_y}$, $(\mathbf{k}^*)^2 := (\mathbf{k}_y)^2 + (\mathbf{k}_z^*)^2$. In the matrix $\mathcal{A}$, the entries of the columns indicated by $j = 2,3,4,5,6$ have the entries as the column with $j = 1$, but with k_z^j instead of k_z^1.

Again, one can show that generically the matrix $\mathcal{A}$ is invertible. The required amplitudes E_z^R and E_z^T are determined by

$$E^R = [(\mathcal{A}^{-1}\mathcal{F})]_1, \quad E^T = [(\mathcal{A}^{-1}\mathcal{F})]_8.$$

We note that for TM polarization the alternative condition (C_5) reads $0 = 0$, while the equations generated by (C_6) on Γ_+ and Γ_0 can be deduced from the equations generated by the main interface conditions and the dispersion relations (62).

The derivation of the Fresnel coefficients is the third major achievement of this work. They are explicitly documented in this work and can be used right away for further research.

VIII. DISCUSSION AND CONCLUSIONS

The aim of this paper was to introduce, on the one hand, a viable route to describe homogenized metamaterials beyond the assumption of a local response and to show the importance of retaining nonlocal material laws in order to adequately describe the dispersion relation of a homogenized metamaterial. Of course, we are not the first to work in this direction, but the approach we have chosen here is generally applicable and does not hinge on the assumption of a specific geometry of the metamaterial. At the example of a fishnet metamaterial, we have shown that WSD is not enough to properly capture the dispersion relation as their functional dependency and their isofrequency contours—either hyperbolic or elliptic—are too simplistic to give accurate predictions beyond the paraxial regime. Significant improvement only comes by introducing nonlocal material laws in their effective description. These come with more degrees of freedom, hence more complicated isofrequency comes into play and gives a better description of metamaterials. In our work, we have studied the light propagation in the bulk of a metamaterial and we obtained all the coefficients that enter the dispersion relation by means of optimization in comparison to the numerically calculated dispersion relation of an actual metamaterial.

Moreover, interface conditions are derived from first principles. They rely on the evaluation of a generalized formulation of Maxwell's equations in a small volume entailing the interface. We have been deriving four main interface conditions and also formulated two alternative conditions. They do not introduce further information but basically look simpler. This might be occasionally beneficial. For a single interface, the consideration of all interface conditions is necessary. This might sound surprising but the nonlocal metamaterial sustains multiple plane waves as eigenmodes at each given frequency. This is in contrast to a local medium where only a single plane wave is supported.

Besides the actual interface conditions, we also derived explicit expressions for the Fresnel equations that can predict reflection and transmission from a slab upon illuminating it with a plane wave. We discuss both TE and TM polarization. The Fresnel equations are documented in a convenient way and are expressed in matrix notation.

With this work, further research endeavours in the context of metamaterials are immediately possible where the physics of such nonlocal metamaterials can be explored. It starts by developing suitable retrieval procedures and the actual quantification of the nonlocality of the metamaterials. It can be extended by analyzing basic optical phenomena in the presence of metamaterials with a strong nonlocality and studying potential applications that rely on such nonlocal metamaterials. Also, the development of suitable numerical tools to explore light propagation in nonlocal metamaterials is an important and timely issue. Finally, based on the general formalism, other kinds of nonlocal constitutive relations can be explored.

ACKNOWLEDGMENTS

We gratefully acknowledge financial support by the Deutsche Forschungsgemeinschaft (DFG) through CRC 1173. K.M. also acknowledges support from the Karlsruhe School of Optics and Photonics (KSOP). C.S. acknowledges the support of the Klaus Tschira Stiftung. We would like to thank R. Suryadharma and A. Vetter for proofreading the manuscript.

[1] A. Alù, Phys. Rev. B **84**, 075153 (2011).
[2] D. R. Smith, Phys. Rev. E **81**, 036605 (2010).
[3] S. Guenneau and S. A. Ramakrishna, Compt. Rend. Phys. **10**, 352 (2009).
[4] M. G. Silveirinha, A. Alù, and N. Engheta, Phys. Rev. B **78**, 075107 (2008).
[5] P. Spinelli, M. Verschuuren, and A. Polman, Nat. Commun. **3**, 692 (2012).
[6] S. Fahr, C. Rockstuhl, and F. Lederer, Phys. Rev. B **88**, 115403 (2013).
[7] A. Alu, F. Bilotti, N. Engheta, and L. Vegni, IEEE Trans. Ant. Prop. **55**, 1698 (2007).
[8] A. Andryieuski, S. Ha, A. A. Sukhorukov, Y. S. Kivshar, and A. V. Lavrinenko, Phys. Rev. B **86**, 035127 (2012).
[9] V. A. Markel and J. C. Schotland, Phys. Rev. E **85**, 066603 (2012).
[10] A. Sihvola, Photon. Nanostructur. Fundam. Appl. **11**, 364 (2013).
[11] M. G. Silveirinha, Phys. Rev. B **75**, 115104 (2007).
[12] X.-X. Liu and A. Alù, Phys. Rev. B **87**, 235136 (2013).
[13] A. Chipouline, C. Simovski, and S. Tretyakov, Metamaterials **6**, 77 (2012).
[14] V. A. Markel and I. Tsukerman, Phys. Rev. B **88**, 125131 (2013).
[15] I. Tsukerman and V. A. Markel, Phys. Rev. B **93**, 024418 (2016).
[16] P. Grahn, A. Shevchenko, and M. Kaivola, Opt. Express **21**, 23471 (2013).
[17] P. Grahn, A. Shevchenko, and M. Kaivola, New J. Phys. **15**, 113044 (2013).
[18] A. Shevchenko, M. Nyman, V. Kivijärvi, and M. Kaivola, Opt. Express **25**, 8550 (2017).
[19] M. Strunc, WSEAS Trans. Electron. **4**, 208 (2007).
[20] J. A. Kong, *Electromagnetic Wave Theory* (Wiley, New York, 1986).
[21] A. Serdyukov, I. Semchenko, S. Tretyakov, and A. Sihvola, *Electromagnetics of Bi-anisotropic Materials: Theory and Applications* (Gordon and Breach, Amsterdam, 2001).
[22] C. Menzel, T. Paul, C. Rockstuhl, T. Pertsch, S. Tretyakov, and F. Lederer, Phys. Rev. B **81**, 035320 (2010).
[23] S. V. Zhukovsky, A. Andryieuski, O. Takayama, E. Shkondin, R. Malureanu, F. Jensen, and A. V. Lavrinenko, Phys. Rev. Lett. **115**, 177402 (2015).
[24] A. Andryieuski, A. V. Lavrinenko, and S. V. Zhukovsky, Nanotechnology **26**, 184001 (2015).
[25] C. Menzel, C. Rockstuhl, T. Paul, F. Lederer, and T. Pertsch, Phys. Rev. B **77**, 195328 (2008).
[26] R. Ghasemi, X. Le Roux, A. Lupu, A. De Lustrac, and A. Degiron, ACS Photon. **2**, 1129 (2015).
[27] A. Ciattoni and C. Rizza, Phys. Rev. B **91**, 184207 (2015).
[28] C. Fietz and C. M. Soukoulis, Phys. Rev. B **86**, 085146 (2012).
[29] T. Geng, S. Zhuang, J. Gao, and X. Yang, Phys. Rev. B **91**, 245128 (2015).
[30] L. Sun, Z. Li, T. S. Luk, X. Yang, and J. Gao, Phys. Rev. B **91**, 195147 (2015).
[31] M. A. Gorlach and P. A. Belov, Phys. Rev. B **92**, 085107 (2015).
[32] M. G. Silveirinha and P. A. Belov, Phys. Rev. B **77**, 233104 (2008).

[33] I. Tsukerman, J. Opt. Soc. Am. B **28**, 2956 (2011).
[34] B. M. Wells, A. V. Zayats, and V. A. Podolskiy, Phys. Rev. B **89**, 035111 (2014).
[35] L. D. Landau, L. P. Pitaevskii, and E. Lifshitz, *Electrodynamics of Continuous Media* (Elsevier, Amsterdam, 2013).
[36] V. M. Agranovich and V. L. Ginzburg, *Crystal Optics with Spatial Dispersion, and Excitons* (Springer-Verlag, Berlin, Heidelberg, 1984).
[37] G. Dolling, C. Enkrich, M. Wegener, C. M. Soukoulis, and S. Linden, Opt. Lett. **31**, 1800 (2006).
[38] F. G. Friedlander, *Introduction to the Theory of Distributions* (Cambridge University Press, Cambridge, 1982).
[39] A. Friedman, *Partial Differential Equations* (Holt, Rinehart and Winston, New York-Montreal Que.-London, 1969).
[40] A. Khrabustovskyi, K. Mnasri, M. Plum, C. Stohrer, and C. Rockstuhl, arXiv:1710.03676.